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Agricultural Sciences

A comparison of two methods to reduce internal phosphorus cycling in lakes: Aluminium versus Phoslock

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Master thesis · Second cycle, A2E

Ultuna, August 2015

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Credits: 30 ECTS

Level: Advanced, A2E

Course title: Independent Project in Environmental Science

Course code: EX0431

Program/education: MSc EnvEuro

Place of publication: Ultuna

Year of publication: 09/2015

Online publication: http://stud.epsilon.slu.se/

Cover picture: yes

Keywords: aluminium, phoslock, phosphorus, lake, restoration, adsorption, treatment

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Aquatic science and assessment

Abstract

Sediment nutrient inactivation through P binding agents is one of the most common management strategies used to prevent potential P release to lake water. Al-based compounds (aluminium sulphate, $Al_2(SO_4)_3 \cdot 18(H_2O)$) and a modified lanthanum-based clay material called Phoslock are among the products that can be used to chemically bind P permanently in sediment. In the present thesis, aluminium sulphate (Alum) and Phoslock were compared in terms of dose application and P binding efficiency in a controlled setting in order to develop dosing models that can be further used in lake restoration and management. Furthermore, a literature review of past and current research was performed to be able to compare both products in terms of potential non-target side effects and treatment longevity. Results showed that Al was able to bind 93-95% of mobile P at the highest dose (Al:Mobile P 150:1). Moreover, cost-effective Al dose ratios were 75:1 for samples with mobile P ranging from 1.55-0.64 mg/g and 25:1 for samples with mobile P ranging from 0.33-0.12 mg/g. By comparison, Phoslock was only able to bind a maximum of 25-35% of mobile P in sediments at the Phoslock:Mobile P dose ratio of 150:1. A comparison between theoretical and real binding ratios also showed that Al was more effective than Phoslock at removing mobile P from sediments. P partitioning after Al treatment showed an increase in the Al-P fraction, whereas Phoslock treatment increased P content in the Ca-P fraction. Treatment costs were lower for Al than for Phoslock and were 83 €/kg of P and 1227 €/kg of P inactivated, respectively. Obtained results, together with potential non-target side effects and treatment longevity gathered from literature, showed that, in general terms, Al is a better product, but that Phoslock may be better in shallower systems with frequent sediment resuspension.

Keywords: aluminium, phoslock, phosphorus, lake, restoration, adsorption, treatment

Contents

Popular summary

Historically, human activity has impacted the environment to the point that damaged ecosystems cannot recover on its own. Eutrophic lakes (lakes with excess of nutrients) are an example of human-induced degradation, mainly because of agriculture and urbanization.

In order to improve lakes' health, nutrient inputs need to be reduced, either if they are originated in the surrounding agricultural fields and lands (external loads) or within the lake (internal loads). In some cases, the reduction of external loads is not sufficient to improve lake water quality; therefore, other approaches need to be considered, for example, acting upon the internal sources of nutrients.

One internal source of nutrients is the sediments of the bottom of the lake, which can release them under certain conditions. One way to avoid such process is to increase the nutrient capacity of the sediments and to strengthen their ability to fix them, so that more nutrients can be stored permanently in sediments. This can be achieved with the addition of chemical agents to lake sediments.

There are several products that can be used but, in this thesis, only aluminium sulphate (Alum) and Phoslock were analysed. The former has been used successfully 30-40 years, whereas the latter is reasonably new (engineered in the mid-1990s) and its properties are still to be completely understood. Both products were applied to sediment samples from Växjö Lake (*Växjösjön*) in order to compare their effectiveness and the doses required to achieve management goals. Furthermore, other non-target side effects that can affect treatment efficiency were also studied. Results showed that Alum is better than Phoslock at fixing phosphorus and that Alum treatments are cheaper. However, and due to its properties, Phoslock could be a better approach in some situations.

Therefore, even though Alum is generally better, product application and treatment choice will always depend on the properties of the lake and those need to be considered if trying to avoid negative non-target side effects.

Summary

Historically, human development has put a lot of pressure on natural resources and many have been damaged up to a point where the ecosystem cannot recover on its own. Lakes and reservoirs, which are not only appreciated for their natural beauty, but also for their contribution to natural resources and services, recreational activities, and other aesthetic purposes, are an example of such degraded ecosystem, mainly due to excess nutrient inputs coming from agricultural activities and urbanization.

Towards the recovery of these water bodies' health, and after the implementation of the Water Framework Directive, a directive from the European Union, great efforts have been carried out to reduce nutrient loads to lakes and reservoirs. Even after the application of management actions at the catchment level, aiming to reduce or eliminate external sources of nutrients, many lakes and reservoirs still present excess of nutrients historically accumulated in waters and sediments, failing to achieve water quality requirements and stressing the need to address such problems.

One way to reduce the presence of dissolved nutrients in waters is to increase the phosphorus adsorption capacity of sediments. By fixing phosphorus in sediments, the nutrient content in solution gets reduced but also its potential release from the sediments into the water. This can be achieved with the addition of binding agents to lake sediments, which have a high phosphorus retention capacity, further leading to an increased nutrient adsorption capacity of sediments. There are several products that can be used as binding agents but, in this thesis, attention was placed upon aluminium sulphate (Alum), which has been successfully used for 30-40 years, and Phoslock, a mid-1990s engineered product that is referred to as a similar or even better alternative to Alum and its use for lake restoration purposes is growing, even though its properties are still to be completely understood.

In the present thesis, both Alum and Phoslock were applied to sediment samples from Växjö Lake (*Växjösjön*) in order to compare phosphorus binding effectiveness of each product, as well as required doses to achieve such phosphorus removal. Furthermore, other non-target side effects that can affect treatment efficiency were also studied. Results showed that Alum was able to bind more than 90% of the available phosphorus in sediments, whereas Phoslock was only able to remove a maximum of 35% of the available phosphorus in sediments. Treatment costs were also considered when assessing cost-effectiveness of each product, resulting in Alum treatments being an order of magnitude less expensive than application of Phoslock (excluding application costs).

Results from the present thesis, together with information of other potential non-target side effects and longevity of treatments gathered from reviewed literature, showed that, in general terms, Alum seems to be a better strategy towards the inactivation of phosphorus and recovery of the lake water quality. However, results also showed that Phoslock can be more appropriate than Alum in certain lake conditions, indicating that product application and choice of treatment also depend on the properties of the ecosystem and that these need to be considered if trying to avoid any negative non-target side effects.

1. Introduction and aim of the thesis

This study focused on lake restoration strategies via reduced internal nutrient cycling between sediments and water in eutrophic lakes. Specifically, two products (aluminium sulphate (Alum) and Phoslock) used to reduce internal phosphorus (P) loading in lakes were studied, aiming to estimate the optimal dose to treat sediments and compare the P reduction relative efficiencies of the two products. In order to do so, a dosing study of both Phoslock and Alum was developed, adding to sediments a range from low to high doses to cover what would likely be both under-dosed and over-dosed treatments. Dosage calculations were based on the bioavailable P load in surficial sediment, which are the releasable P fractions (i.e., mobile P). Therefore, the mass of product to mobile P mass was used to estimate how much of the binding agent was needed for each sediment sample. Furthermore, a review of past and current study cases was also carried out to assess other factors affecting binding effectiveness and complement the results presented in this thesis.

According to other research studies, Al (Alum) is a compound that has been used in the last 30-40 years for lake restoration purposes (and even longer for water treatment) and has given great results in terms of increased P adsorption in sediments and, thus, effective removal of P in lake water. On the other hand, Phoslock is a newly developed product that is currently advertised as an equivalent or even better alternative to Al and its use in lake restoration projects is growing, even though its characteristics and performance are still to be completely understood. The hypothesis of the current study was that both Phoslock and Al would be able to achieve similar reductions in mobile sediment P (via conversion to more inert forms), and thus the costs for application would be similar as well.

The results of this study will help estimate how much Al or Phoslock needs be added to bind a certain amount of mobile P in the sediment.

2. Basics of lake restoration

Lakes and reservoirs are not only appreciated for their natural beauty, but also for their contribution to natural resources and services, recreational activities, and other aesthetic purposes. Human development has put a lot of pressure on them and many have degraded due to excessive nutrient inputs.

Accumulation of nutrients coming from anthropogenic sources can worsen the ecological structure and function of lakes, affecting the ecosystem services provided by them and enhancing algal blooms and eutrophication processes, which are thought to intensify due to climate change (Jeppesen et al. (2000), Paerl and Huisman (2008), Feuchtmayr et al. (2009), Jeppesen et al. (2009), Smith and Schindler (2009), Brookes and Carey (2011)).

Since the implementation of the Water Framework Directive (WFD), many efforts have focused on the reduction of external nutrient sources to lakes. However, the “good ecological status” target for all lakes by year 2020 stresses the need to manage, as well, internal nutrient loadings from sediments so that water quality goals are met (Mackay et al. (2014), Spears et al. (2014)).

2.1. Limnology of lakes

Deep lakes are characterized by thermal stratification (also called dimictic lakes), with the warmer zone in the upper part during summer but inverse order during winter. This process is controlled by wind, solar input, and water density variation due to temperature (Cooke et al., 2005). In turn, thermal stratification isolates the deeper waters from the atmosphere, limiting light penetration and generation and diffusion of oxygen. Consequently, respiration processes in such conditions lead to dissolved oxygen depletion, creating reducing conditions and resulting in nutrient release from sediment iron complexes (Caraco et al. (1989), Golterman (1995), Gätcher and Müller (2003)). By comparison, spring and fall are periods of continuous mixing across the water column (Wetzel, 2001). Deep lakes have a smaller surface area exposed to wind mixing (relative to lake volume), which, together with stratification, provide a great resistance to vertical transport of nutrients to superficial waters (Osgood, 1988). This is the reason why these systems show nutrient-limited algae growth in surface waters. By comparison, internal nutrient release processes in a lake with low resistance to mixing and vertical transport (shallow lakes, for example) can stimulate algal blooms and eutrophication (Stauffer and Lee (1973), Larsen et al. (1981), Cooke et al. (2005)).

In shallow lakes (mean depth ≤ 3m), mixing processes happen continuously and stratification periods only occur briefly after calm and hot weather periods (also known as polymictic lakes) (Cooke et al., 2005). Thereby, day/night cycles of high/low concentrations of dissolved oxygen stimulate nutrient release during night, especially below macrophyte beds (Frodge et al., 1991). Macrophytes (rooted, emergent, floating and submersed plants) are the typical vegetation in shallow lakes (or littorals of deep lakes) and their presence depends on light availability (Canfield et al. (1985), Barko et al. (1986), Duarte and Kalff (1986) (1988), Smith and Barko (1990)). In turn, they stabilize and compact bottom lake sediments, reducing resuspension and internal nutrient loadings (Bachmann et al. (2000), Anthony and Downing (2003), Horppila and Nurminen (2003)). In these lakes, bottom-surface interactions affect the whole water column; therefore, internal loadings will increase nutrient levels in the lake (Cooke et al., 2005).

2.2. Eutrophication and nutrient cycling in lakes

Eutrophication is caused by inputs of organic and inorganic matter to lakes at rates that are sufficient to increase biological production, leading to nutrient enrichment of sediments (due to organic matter mineralization processes), loss of lake volume, and depletion of dissolved

oxygen. Nutrient availability can also increase through respiration processes stimulated by organic matter (Rydin, 1985). Moreover, energy from organic matter also increases living biomass (Wetzel (1995) (2001), Cole (1999)). Likewise, silt loadings may also contribute to nutrient release and volume loss, the latter enhancing the development of shallow areas in lakes (Carpenter (1980) (1981) (1983)).

Eutrophic lakes are characterized by floating microscopic cells (planktonic), presence of blue-green algal blooms (cyanobacteria) and of macrophytes, and coloured water (green/brown) (Cooke et al., 2005). Consequently, aquatic climates become turbid and light-limited, leading to low dissolved oxygen levels in the deeper parts and restraining primary production, biomass, and areal distribution of rooted vegetation (Egemose et al., 2010).

In light of climate change, warm lake waters also contribute to algal blooms via faster algal growth. Moreover, zooplankton's sensitivity to high temperatures and the higher presence of *warmwater* fish, which feed from zooplankton, further decrease control on algae (Jones and Hoyer (1982), Cooke et al. (2005)). In addition, warmer sediments allow higher microbial decomposition rates and nutrient release (Jensen and Andersen, 1992).

Nutrient loadings to sediments come from organic matter, soluble P, and other inorganic P forms that enter from external sources or are produced in the lake. External nutrient loadings to lakes may be, during the warmer periods of the year, lower than internal cycling of sediments. Internal loading in lakes can be caused by biological processes, such as sediment disturbance by release of methane gas or burrowing animals, microbial activities, or temporary anoxia. Likewise, chemical processes (e.g., increase in pH due to photosynthesis, lack of oxygen and low redox potentials) and physical processes (e. g., wind induced turbulence) can also cause nutrient recycling (Cooke et al., 2005). Due to the nature of those processes, productivity is negatively correlated with mean depth, therefore, shallow lakes will present more macrophytes than deep lakes (Wetzel, 2001).

Elevated sediment mixing caused by, for example, carp (Huser et al., in press) may lead to increased availability of sediment P, feeding phytoplankton blooms and resulting in the worsening of the light climate of the aquatic ecosystem, which in turn hinders the settlement of rooted vegetation. Wind exposed lakes and shallow lakes are usually the most sensitive to resuspension, the latter due to the sediment's properties, which are not optimal for sediment consolidation (Kristensen et al., 1992). In addition, not compacted sediments work against biological stabilization, as rooted vegetation lacks anchoring opportunities (Schutten et al., 2005).

2.2.1. Phosphorus in lake sediments

The control of algal biomass in the long term requires nutrient reduction in the water column. Towards this goal, P is usually the most targeted nutrient for different reasons. First of all, because P is the limiting factor for algae production (Schindler et al. (2008), Welch (2009)). Moreover, N can be fixed by cyanobacteria and controlled by microbial activity, which affect both nitrification and denitrification processes at the sediment-water interface (McCarthy et al. (2007), Gibbs and Özkundakci (2011)), whereas there are no similar controls for P. In addition, P does not contribute in gaseous loadings, unlike N or C, hence, lake P content can be easily controlled by managing external and internal loadings (Cooke et al., 2005). Therefore, it is reasonable to conclude that internal sources of N are more difficult to control than P internal loadings.

P bound to sediments is distributed in the form of different fractions (labile, Fe-P, Al-P, Org-P, and Ca-P). Consequently, P compounds sharing similar reactivity will be clustered in the same fraction and will be released under specific conditions, different from the other fractions. The

labile fraction corresponds to the immediately available P (water soluble, dissolved, or loosely bound to surfaces of Fe and $CaCO_3$); the Fe-P fraction refers to the reductant soluble P, which is mainly bound to Fe-hydroxides and Mn-based compounds; the Al-P fraction comprises the P that is bound to metal hydroxides of Al (inorganic and soluble in bases); the Org-P fraction corresponds to the P that is bound to organic matter (microorganisms, detritus, humic compounds, etc.); and the Ca-P fraction refers to the more inert P form, also known as "*apatite bound P*" (bound to carbonates and apatite) (Psenner and Pucsko (1988), Hupfer et al. (1995)).

In general, labile and Fe-P are regarded as more reactive and bioavailable, from which internal P loadings are mostly released to the water column. Pore-water and loosely bound P (labile fraction) is generally available and released under all conditions, whereas Fe-P is mainly released due to the mineralization of the organic matter, causing oxygen depletion and periods of water anoxia (Smolders et al., 2006). Sediment P released under anaerobic conditions mainly comes from the Fe-P fraction, where the nutrient is bound to Fe(III) and Mn(III/IV) compounds and it is released after reduction to Fe(II) and Mn(II) (Boström et al., 1988). On the contrary, Al-P, Org-P and Ca-P fractions are regarded as more refractory or inert. Nevertheless, Al-P can be released, together with Fe-P, under sustained low and high pH levels (i.e., below 5.5 and above 9), Org-P labile forms can degrade over time and become bioavailable, and Ca-P can be released at lower pH levels (solubility starts to increase at pH <8).

The magnitude of internal P cycling between sediment and the water column depends on the site's physicochemical properties (pH, redox conditions, wind induced sediment resuspension, etc.) (Boström et al. (1982), Boström et al. (1988), Lukkari et al. (2007), Spears et al. (2011)), the amount of releasable P in the sediment (Huser and Pilgrim 2014), and other biological factors (microbial activity, macrophyte cover, etc.) (Boström et al. (1982), Boström et al. (1988), Phillips et al. (1994), Stephen et al. (1997)). Nevertheless, estimates for such P release depth in sediments generally range between 4 and 10 cm (Boström et al. (1982), Cooke et al. (2005)).

In the present study, mobile P, considered the potentially releasable sediment P pool ((Søndergaard et al. (2003), Pilgrim et al. (2007)), was assumed to be the sum of the labile and Fe-P fractions, because they are the most reactive and bioavailable. The Org-P fraction was not included in the mobile P pool because it requires time to mineralise and convert to mobile P, and it would not make sense in the present short-term laboratory settings. Furthermore, at a single point in time and after the degradation of Org-P and transformation to mobile P, a "new" Org-P fraction will exist due to the continued deposition of phytoplankton and of decomposing macrophytes (Meis et al., 2013). Therefore, the actual/measured Org-P fraction will not be available, only the mobile P (labile and Fe-P), which is very important in terms of treatment dosages and lake restoration. An actual dose calculation for lake management, however, should consider the labile Org-P fraction.

2.3. Management of P in lakes

In the last few decades, nutrient management programs have aimed to control and reduce nutrient loads, coming from surrounding ecosystems, to lakes and other water bodies (Carpenter (2008), Schindler et al. (2008)). Minimization of external P sources has been achieved with actions at the catchment level, such as the enhancement of riparian buffer zones, reduction of point discharges, restoration of wetlands around the lake shore, or the control of lake and river inflows (Gibbs et al., 2011). Nevertheless, even after a successful reduction of external P loads, lakes often still have elevated P concentrations due to the nutrient cycling between the water column and lake sediment (also called internal P loading), which can hinder the recovery of the lake (Marsden (1989), Vyhnâlek (1991), Søndergaard et al. (2003), Jeppesen et al. (2005)). Moreover, in some occasions, internal loadings can be higher than external loadings, which emphasize the fact that management of external sources of P may not

be sufficient to eliminate eutrophication and achieve good water quality. In addition, actions at the watershed level may require more infrastructures and may be more difficult to implement than methods aimed at internal nutrient loadings.

Various techniques have been tested aiming to minimize internal nutrient cycling in eutrophic lakes (Cooke et al. (2005), Hupfer and Hilt (2008), Hickey and Gibbs (2009)). Some of these methods include sediment removal (Hupfer and Hilt, 2008), hydraulic flushing (Hosper and Meyer, 1986), and sediment capping, which can be done with modified local soils (Pan et al., 2012), with calcite (Berg et al., 2004), modified clay minerals (Robb et al., 2003), or the addition of iron slag (Yamada et al., 1987). P stripping agents (Gibbs et al., 2011), which increase the P adsorption potential of the sediment allowing a higher nutrient retention, are also used. Most common agents are iron ($FeSO_4$, $FeCl_3$) (Perkins and Underwood (2001), Boers (1991)), Al ($Al_2(SO_4)_3$, $AlCl_3$, Al modified zeolite) (Welch and Cooke (1999), Lewandowski et al. (2003a), Reitzel et al. (2005), Özkundakci et al. (2010)), calcium ($CaCO_3$) (Hupfer and Hilt, 2008), and lanthanum based products (Phoslock) (Gibbs et al. (2011), Meis et al. (2012)). The natural product Allophane (Yuan and Wu, 2007) and a modified zeolite product called Aqual-P (Z2G1) (Gibbs and Özkundakci, 2011) are also among the products that have been tested for their P binding potential. Finally, artificial oxygenation of sediment iron has also been trialed (Gächter and Wehrli, 1998).

Loads of accumulated P in lake sediments can also be released through resuspension during warm weather seasons (Vyhnâlek, 1991). The reduction of resuspension events, together with the inactivation of sediment P, would enhance benthic production, oligotrophic conditions, and consolidation of sediments (Bengtsson et al., 1990). In turn, sediment stabilization can reduce or even prevent resuspension, which is the most common pathway of P release (Egemose et al., 2009). Therefore, all these factors need to be considered when applying chemical treatments for lake restoration.

2.3.1. Aluminium (Al)

Aluminium sulphate (Al, $Al_2(SO_4)_3 \cdot 18(H_2O)$) is, currently, one of the most used P inactivation agents. Nevertheless, depending on the properties of the lake and the relative magnitude of P sources, its effectiveness can be limited or its application even not appropriate.

Geochemistry

At pH 6-9, Al added to water produces a gelatinous/amorphous precipitate ($Al(OH)_3$) that is stable enough to face the anoxic environments of summer lake stratification and provide long-lasting binding sites for P (PO_4^{-3}) (Gibbs et al., 2011). During the formation of such precipitate, hydrogen (H^+) ions are released due to a hydrolysis reaction, consequently lowering the pH of the water if adequate lake water alkalinity is not present. If a buffering compound is not used in low alkalinity systems, pH depression can lead to formation of soluble and toxic Al^{+3} and $Al(OH)_2^+$ ions and poor sediment particle aggregation which, in turn, enhances resuspension events (Afsar and Grooves (2009), Egemose et al. (2010)). Therefore, water alkalinity will limit the maximum dose of Al that can be applied without lowering the pH too much unless buffering compounds (sodium aluminate ($NaAlO_2$) or lime ($Ca(OH)_2$), for instance) are used (Cooke et al., 2005).

In addition, lake water with sustained (>12 hours) high pH levels (pH>9.5) due to photosynthesis may increase the risk of releasing P from Al hydroxides in sediments (Rydin and Welch, 1999) and reduce Al sorption efficiency due to P substitution by hydroxyl anions (Reitzel et al., 2013). pH is typically 1-2 units lower near the sediment surface compared to the epilimnion (surface waters), however, and pH after a restoration should not be elevated (except

in very alkaline lakes) due to limitation of productivity and CO_2 consumption by algae (Huser et al., 2011).

Response to treatment

Al application increases the P adsorption capacity of sediments (Rydin and Welch (1999), Lewandowski et al. (2003b), Reitzel et al. (2005)). After properly dosing Al relative to mobile P, a general reduction of P in lake water and a 90% decrease of internal P loadings are observed (Reitzel et al., 2005). Furthermore, Al treatments can achieve P reductions greater than 90% (Pilgrim et al., 2007).

Adsorption of P by the Al precipitate ($Al(OH)_3$) may be limited by interactions with other compounds that are also present in the water column and compete for the available binding sites. Al complexation with sulphates and fluorides (Hem and Roberson, 1967), phosphate (Guan et al., 2006), organo-Al complexes (Bloom, 1981), and organic matter (Lind and Hem, 1975) could limit P sorption in lake sediments, but the latter seems to be the most important and probable interference (Hessen and Tranvik, 1998). Furthermore, silicates and dissolved organic carbon can also compete with P for binding sites of the Al precipitate (de Vicente et al., 2008).

Other factors could also affect the sorption efficiency, such as lake morphology and Al dose (Huser, 2012), $Al(OH)_3$ aging (crystallization) and mobility (de Vicente et al., 2008), resuspension events, localized P limitation, benthic disturbances, or other chemical changes in the Al precipitate or lake water (Huser et al., 2011). Focusing of Al (Al accumulation in excess of mobile P) also reduces Al sorption efficiency due to increased resuspension potential and translocation of Al from shallow to deeper parts of lakes, where P may be limited (Egemose et al. (2010), Huser (2012)).

Al binding efficiency of P has been shown to differ in previous research and lake restoration projects, and Al added to Al bound P (Al:Al-P) molar ratios can range from 2.1:1 to 14.5:1 by weight (Rydin and Welch (1999), Rydin et al. (2000), Lewandowski et al. (2003b), Reitzel et al. (2005), Huser et al. (2011), Huser (2012)).

After Al treatment, Al-P is the only fraction that shows an increase in its content, thereby, mobile P is converted to Al-P, and the produced $Al(OH)_3$ precipitate is recovered from the corresponding extract of the sediment fractionation ($NaOH{\cdot}P$, see section 3.4) (Rydin and Welch (1999), Hansen et al. (2003), Reitzel et al. (2005)).

When Al is applied to restore lakes, an increase of the Al-P fraction is also observed over time. This is due to newly adsorbed P from new settling sediments or organic matter and from the sediments within and below the Al layer. Therefore, Al also works against diffusion of P from deeper sediments while there still is excess binding capacity (Reitzel et al. (2005), Huser (2012)). However, Al aging reduces maximum binding capacity by up to 75% in absence of P (de Vicente et al., 2008). Longevity and effectiveness of Al treatments has been found to be highly variable and beneficial effects can last from <1 to more than 40 years (Welch and Cooke (1999), Huser et al. (2015)). Nevertheless, dosing repetition over time can improve Al efficiency (Lewandowski et al. (2003b), de Vicente et al. (2008)).

Al treatment might have limited effectiveness in lakes with a high amount of wind induced mixing and high pH, due to the potential for dissolved Al release in the anionic form (Reitzel et al., 2013). Moreover, Al can get buried under re-settling sediments, due to considerable Al mobility (Huser, 2012). Also, other effects after treatment include a high sedimentation rate, an improved and immediate P uptake, and a high efficiency in removing colour and improving light conditions (Egemose et al., 2009).

Dosage in other case studies

Even though, for a time, Al dosage was based on lake water alkalinity (Kennedy and Cook, 1982), current methods dose Al based on the actual mobile P in the sediment that contributes to internal P cycling (Rydin and Welch, 1999).

A solution of Al in deionized water, buffered to keep pH levels around 6, with a final Al concentration of 0.7 g/l was used by Rydin and Welch (1999) to treat sediments with a Al:Mobile P ratio of 100:1 (by weight). Another suggested Al to mobile P ratio was 10:1 (molar, 11:1 by weight) (de Vicente et al., 2008), but neither of these were chosen according to system conditions, therefore, they could result in the under or over dosage of mobile P, and even a reduced efficiency of the Al treatment.

More recent research has shown that the required ratio to fix the 90% of the releasable P pool is variable and decreases with increasing mobile P (from 100 to less than 20:1) (James, 2011). A molar Al:Mobile P ratio of 4:1 (equivalent to 3-4:1 ratio by weight) was used by Reitzel et al. (2005), and a ratio of 10:1 by Egemose et al. (2010). Huser and Pilgrim (2014) used a set of solutions of aluminium sulphate ($Al_2(SO_4)_3 \cdot 18(H_2O)$) with final Al concentrations ranging from 250 to 850 mg/l to achieve Al:Mobile P ratios of 25, 50, 75, 100, 125, and 150:1 (by weight) based on initial mobile P content. Models that can predict the reduction of mobile P in the sediment with a specific dose were then developed, allowing for a more flexible determination of Al dose to reduce internal P release from sediment.

2.3.2. Phoslock

Phoslock is an alternative to Al that is rapidly emerging as a solution for lake restoration but it is still not clear whether it offers an improvement or not. Phoslock is a lanthanum-based (5% La, 95% clay) engineered product developed and patented by the Australian national science agency, the Commonwealth Scientific and Industrial Research Institution (CSIRO), in the 1990s (Finsterle, 2014). Besides La, Fe and Al are also present in the Phoslock matrix, confirming the existence of binding sites other than La (Reitzel et al., 2013).

Phoslock has been used in lake restoration for over 10 years now and is reported to be one of the most effective P inactivation strategies (Finsterle, 2014), although its P binding mechanisms are not completely understood and need further investigation, especially in terms of Phoslock performance depending on lake water conditions. Several publications report on Phoslock efficiency, but not all of them reach similar conclusions. In some cases, Phoslock applied to sediments worked poorly (Meis et al. (2012), Meis et al. (2013)), whereas other studies obtained significant P removal efficiencies in solution (Robb et al. (2003), Ross et al. (2008), Vopel et al. (2008), Haghseresht et al. (2009), Geurts et al. (2011), Gibbs et al. (2011), and Reitzel et al. (2013)).

Geochemistry

Phosphate (PO_4^{-3}) precipitation by lanthanum (La^{+3}) (highly stable mineral called *Rhabdophane*, $LaPO_4$) is effective at a wider pH range compared to Al and Fe (4.5-8.5) and is produced at low pH, when it is also highly insoluble (Haghseresht et al., 2009). A pH increase leads to higher content of PO_4^{-3} (due to loss of hydrogen ions), but also to hydroxylation of La ions ($La(OH)_3$), reducing P uptake capacity of Phoslock (Haghseresht et al., 2009). In addition, the pH effect in Phoslock efficiency is more important in high alkalinity than low alkalinity waters because of a greater CO_3^{-2} formation, which become a competitor with P for La binding sites (Lürling and Tolman, 2010). When it comes to P adsorption, such competition for La binding sites seems to be a more relevant factor, in high alkalinity waters and high pH, than hydroxylation of La (Reitzel et al., 2013). Furthermore, pH effects can be reversible, improving P adsorption to Phoslock by

reducing the pH of the lake water. However, the reversibility would not work both ways and adsorbed P could be released with a pH increase (Reitzel et al., 2013).

Besides the effects that lake water conditions can produce to Phoslock efficiency, Phoslock addition can also induce changes in lake water properties, such as alkalinity, which shows a small increase (from 40.5 mg/l $CaCO_3$ to 47.5 mg/l $CaCO_3$, for example) (Reitzel et al., 2013). On the other hand, Phoslock does not affect lake water pH (Reitzel et al., 2013).

Phoslock adsorption kinetics in natural waters show the highest P uptake occurring within the first 50 minutes, with considerably lower P removal rates after that. In the pH range of 5-9, where P binding to Phoslock is higher, maximum adsorption (93%) is reached at pH 6 and efficiency drops to 81% at pH 9 (*Figure 1*) (Zamparas et al., 2015).

Phoslock dispersion (smaller particles) and, thus, P adsorption, is improved by high alkalinity conditions and is not affected by increasing conductivity. However, Phoslock particle size increases at pH above 8, leading to increased rates of sedimentation and lower P adsorption. Interfering substances (i.e., Ca^{+2}, humic compounds) can reduce Phoslock particle dispersion and decrease La availability for P uptake (Tang and Johannesson (2003), Sonke and Salters (2006), Ross et al. (2008), Tang and Johannesson (2010), Reitzel et al. (2013)). Complexation of La with humic acids can also reduce binding capacity of Phoslock (Lürling and Faassen, 2012), and similar trends have been observed in pore water of peats (Geurts et al., 2011). Therefore, lakes with high levels of humic or dissolved organic matter may show reduced P adsorption due to these interactions.

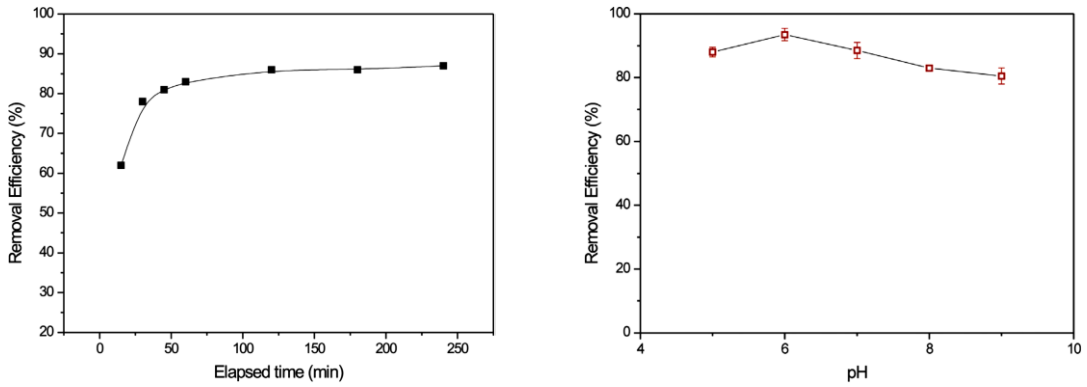

Figure 1, Adsorption kinetics and influence of pH on P uptake by Phoslock (Zamparas et al., 2015)

Finally, bioturbation activities are greater under aerobic conditions, which lead to improved mixing of Phoslock and sediments, the burial of the Phoslock layer under fresh sediments, and a conductivity increase (Meis et al., 2013).

Response to treatment

Adsorption efficiency of Phoslock in water has been estimated, approximately, at 87% of P removal (Zamparas et al., 2015). It is suggested that relative adsorption of mobile P to Phoslock declines with increasing dosage relative to mobile sediment P (Reitzel et al., 2013). In addition, binding ratios (by weight) have been estimated to range from 9.14 to 8.56 mg P/g Phoslock at ~100:1 Phoslock:Mobile P doses in lake water, whereas maximum binding ratios (11.3-13.7 mg P/g Phoslock) were achieved at a 11:1 Phoslock:Mobile P ratio (Reitzel et al., 2013). Around 21% of P bound to Phoslock is suggested to be releasable, whereas around 79% is unlikely to become available under reducing conditions or common pH (5-9) (Meis et al., 2012).

After Phoslock application, P partitioning and behaviour in sediments has been reported to be different than after Al treatments (see section 2.3.1). P content increases over time in the Ca-P fraction (Meis et al., 2013), which agrees with Meis et al. (2012) in that, approximately, the 60% of the P fixed by Phoslock is found in Ca-P fraction. On the contrary, other sediment P fractions do not show any significant differences after Phoslock application (Meis et al., 2012), even though the small amount of P transferred to the Ca-P fraction seems to come from mobile P (mainly Fe-P fraction) (Reitzel et al., 2013). However, such P increases in the Ca-P fraction do not happen immediately, it occurs gradually over time, which emphasises the importance of Phoslock aging and its efficiency variability, but not enough research has been done in this area (Reitzel et al., 2013). Due to the adsorption time lag that has been detected between Phoslock application and increase of Ca-P, it is suggested to plan Phoslock treatment during winter in order to minimize mobile P release during summer and autumn (Meis et al., 2013).

Phoslock application leads to a significant La content increase and, consequently, to a theoretical increase of P adsorption capacity of sediments due to greater La-P binding sites (Meis et al. (2012), Meis et al. (2013)). In addition, added mass of La has an estimated potential to bind mobile P of 25% (in the top 4 cm) or 10% (in the top 10 cm) (Meis et al., 2013). But La can also move vertically through the sediment, reducing efficiency of Phoslock if it is transported below the mobile P cycling depth. Transport processes that can cause La mobilisation include wind induced resuspension (Hilton et al. (1986), Douglas and Rippey (2000)) or bioturbation (Fisher et al. (1980), Meysman et al. (2006), Reitzel et al. (2013)).

La is associated to particles instead of being dissolved (Reitzel et al., 2013), which is largely positive since La toxicology is attributed to its dissolved form (La^{+3}) (Peterson et al., 1976). In addition, the high pH (>9) or alkaline conditions that usually prevail in eutrophic lakes inhibit La^{+3} ions, minimizing the toxicity risks associated with Phoslock in hard water lakes (Lürling and Tolman, 2010). Nevertheless, some studies have reported leakage of La ions due to Phoslock dispersion (Gibbs et al., 2011).

The effects of resuspension events after Phoslock treatment are, on one hand, a better consolidation and compaction potential of sediments, thereby increasing the erosion threshold, but also a net accumulation of P, meaning that Phoslock does not bind it immediately (Egemose et al., 2009).

Dosages in other case studies

Generally, dosage and addition/application of Phoslock is performed relative to the content of mobile P (sum of labile P, Fe-P, and Org-P) in the sediment (Reitzel et al. (2005), de Vicente et al. (2008)), as it is considered to be adequately measurable with standard sequential extraction techniques (Psenner et al. (1988), Hupfer et al. (1995), Paludan and Jensen (1995)).

The theoretical binding capacity of Phoslock to P is 100:1 (Finsterle, 2014), which is what calculations are usually based on. Nevertheless, as with Al, dosage of Phoslock depends on the characteristics of the experiment or the properties of the lake, and system conditions can affect the adsorption greatly. A Phoslock:P ratio of 100:1 (based on mobile P in the water column and the upper 4 cm of the sediment) was used by Meis et al. (2012) in a treated shallow reservoir in Scotland, whereas Egemose et al. (2010) applied a Phoslock:P ratio of 230:1 (by weight) in their research. Lürling and Faassen (2012) and Reitzel et al. (2013) stated that the 100:1 Phoslock:P ratio they used was not sufficient to adsorb the total mobile P concentration in their studies.

Reitzel et al. (2013) prepared a continuously mixed stock solution of 600 mg/l of Phoslock that was later applied in a 100:1Phoslock:P (w/w) ratio to pore water concentrations of P that were representative of eutrophic lakes (620 µg/l). The treated solution was incubated for 12 days

before analysing. Phoslock:P ratios ranging from 0 to 217 (w/w) were also used to determine whether Phoslock efficiency was affected by water alkalinity and conductivity conditions. A different slurry (60 mg/l Phoslock, 230:1 Phoslock:P ratio, incubated for 24h) was prepared and used to test whether Phoslock itself affected water pH and alkalinity. Meis et al. (2013) prepared a slurry containing 25,000 kg of Phoslock that was applied to a lake, for 3 days, to control 279 kg of P, resulting in a Phoslock:P ratio of 90:1, approximately (24h to let Phoslock settle).

The present study focuses on both Al ($Al_2(SO_4)_3$) and Phoslock, since they currently are two of the most used products to inactivate P of bottom lake sediments. A dosing study of both products is developed, aiming to estimate the optimal dose to treat sediments and compare the relative efficiencies of each of the treatments.

3. Materials and methods

3.1. Study site

Sediment samples were collected from Växjö Lake (*Växjösjön*, 56.86796ºN, 14.81025ºW), located in the municipality of Växjö, in southern Sweden. Växjö Lake is part of the Mörrum main basin and is drained by the Bergunda canal, which runs to Bergunda Lake (*Bergundasjön*), located south-west of Växjö Lake. In addition, Trummen is located south-east of Växjö Lake, whereas Lake Helga (*Helgasjön*) can be found north of the city of Växjö, and all four lakes have been important for the lake water management of the area over the years. Växjö Lake has a surface area of 79 ha, a perimeter of 4600 m, and a mean and maximum depth of 3.9 and 7.8, respectively. Its volume is of 3.1 million m^3 and the annual runoff is of 4.3 million m^3. The outlet of the lake is of 19.6 km^2 (ALcontrol Laboratories, 2013a)

Eutrophication started in Växjö Lake during the 1700s and 1800s due to increasing population in the area and large amounts of untreated waste and wastewater dumped into the lake. The lake exhibited a highly nutritious nature, including occurrence of cyanobacteria during summers, lack of oxygen during winters causing, sometimes, fish deaths, and development of plankton blue-green algae (ALcontrol Laboratories, 2013a). Actions were taken along the years to improve the lake's conditions (instalment of sewage systems, transfer of water from surrounding lakes, nutrient-rich sediment dredging, reduction of pollution sources, water diversion), but they didn't solve the problem, even intensified it for some time. During the 1950-1970s, management in Trummen led to a reduction of P levels in Växjö. Nevertheless, by 1980s, eutrophication in Växjö Lake was not yet eliminated due to continuing P loads from lake's internal cycling (especially in summer) and storm water flows. In the 1990s, fishing activities were increased in order to restore Växjö Lake's ecosystem balance, prior to undergoing sediment dredging processes. Further management efforts achieved the lowest P concentrations ever measured of 26 µg/l (2010-2012) (ALcontrol Laboratories, 2013a).

The lake is currently regarded as a well-functioning lake with public beaches. However, Växjö Lake is characterized for having internal P release from sediments when conditions are not favourable, significant and large presence of blue-green algae, skewed fish composition and a high zooplankton predation from fish, and for not having a large amount of aquatic plants (ALcontrol Laboratories, 2013a). The hydrological load to the lake comes from natural land flows, storm water flows, direct rainfall on the lake surface, and Lake Helga's water transfer. External P loads coming from the entire lake's inflow have been significantly reduced over the last years, from values of 440 kg/year in 1979 to 180 kg/year in 2012, despite the 50% increase of the hydrological load. The reduction of the external load of P also led to the reduction of the nutrient outflow and the internal load, which resulted, in turn, in a P retention rate (negative internal load) of 30 kg/year (2010-2012) (ALcontrol Laboratories, 2013b).

Calculations showed that long-term concentrations for lake water P would be 18 µg/l at current hydrological and external P loads, which would be higher than the target of "good ecological status" (16 µg/l). A large reduction of the external load, together with an increased water transfer from Lake Helga, would achieve the target concentration if performed together with management of the internal loads (by increasing P retention on sediments) (ALcontrol Laboratories, 2013b).

3.2. Sample collection

Three sediment cores were collected at 3 different sample sites and depths (V1 (7.9m), V2 (5.7m), and V3 (1.8m)), approximately 1 month before starting the laboratory experiment, using a *Willner* gravity sediment-coring device. Cores were extracted and sliced (2 cm) to a sediment

depth of 36 cm on site, resulting in a total of 36 sediment samples, and stored at 4ºC in opaque containers (fridge) until preparing them for the analyses.

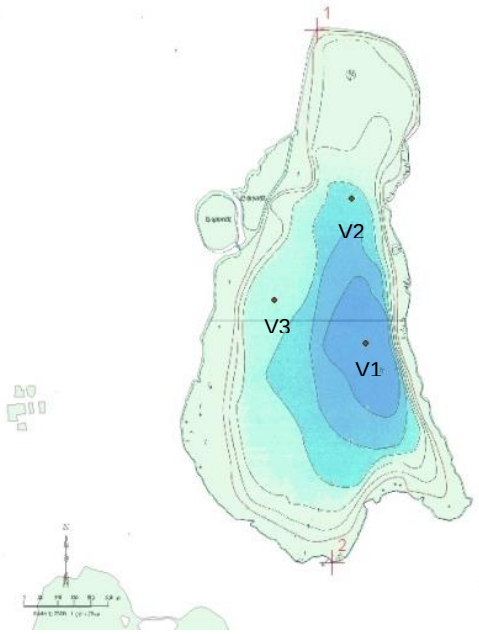

Figure 2, Sampling points (V1, V2, V3) and depth map (colours) for Växjö Lake.

3.3. Basic physical properties of sediments

3.3.1. Water content

Sediment dry weight and water content were estimated for all collected samples (n=36). Wet sub-samples were weighted and frozen at -19ºC for 24h before freezer-drying for 4 days. After that, all sub-samples were placed in the oven at 50ºC for 24 hours. Dry sub-samples were later weighted, and dry weight and water content were calculated as:

$$\%wc = \frac{wet-dry}{wet} \cdot 100 \qquad \%dry = 100 - \%wc$$

where *%wc* equals to the water content (%) of the sediment, *wet* refers to the wet weight of the sub-sample, *dry* equals to the dry weight of the sub-sample, and *%dry* refers to the percentage of dry substance in the sediment.

3.3.2. Density

Density of each layer of sampled sediments (in g/cm^3) was calculated from the water content and following the expression below (unpublished data), based on Håkanson and Jansson (1983).

$$\rho = -0.5923 \cdot ln(\%wc) + 3.7099$$

In addition, density was also estimated after loss on ignition (LOI) (Håkanson and Jansson, 1983). Sub-samples of dry sediments (~200 mg) were weighted and placed in the oven, overnight, until they reached a temperature of 550ºC and were burned for 2h. Burned sub-samples were later weighted and density was calculated based on organic content and water content, as follows

$$\%LOI = \frac{dry-burned}{dry} \cdot 100 \quad \rho = \frac{260}{100+1.6\cdot\left(\%wc+\frac{\%LOI}{100}\cdot\%dry\right)}$$

where *%LOI* equals to loss on ignition, *burned* refers to the weight of the sub-sample after burning, and ρ equals to the estimated density (g/cm^3).

3.4. Sediment analysis

Fractionation techniques are widely used to determine concentrations of specific compounds in sediments according to their chemical behaviour. This method clusters compounds of similar reactivity in different fractions, and the properties of each fraction describe the sediments they are bound to. In general terms, P fractionation separates the P which is already dissolved, from the potential soluble at certain conditions (low redox potentials, high pH, microbial activity, etc.), from the inert fraction. Thus, the total P of the sample is the sum of the P in each fraction.

The sequential extraction technique used in this project is a modification of the scheme for wet sediments described by Hupfer et al. (1995) and based on Psenner et al. (1988). All extracts were centrifuged at 3000rpm during 10 minutes before the analysis with spectrophotometer as soluble reactive P with the molybdate blue method (Murphy and Riley, 1962). The extraction of the different P fractions (labile P, Fe-P, Al-P, Org-P, and Ca-P) was performed with a series of different extractants, including double-deionized water, bicarbonate-dithionite (BD), NaOH and HCl (*Table 1*). For the Al-P fraction, an additional centrifugation was applied in order to minimize organic matter interference in the extract and improve P content analyses. Nevertheless, measurements were recorded before and after the additional centrifugation to compare the results and evaluate the differences.

Table 1, P fractionation scheme used for sediment analyses described by (Hupfer et al., 1995) and based on (Psenner et al., 1988).

Step	***Extractant***	***Time (h)***	***Fraction***	***Org-P***
1	Double-deionized water (H_2O·P)	2	Labile P	Calculated as the difference Tot-P – Al-P
2	$NaHCO_3$ 0.1M / Na_2SO_4 0.1M (BD·P)	1	Fe-P	
3	NaOH 0.1M (NaOH·P)	16	Al-P	
4	NaOH 0.1M digested with $K_2S_2O_8$ (NaOH·P)		Tot-P	
5	HCl 0.5M (HCl·P)	16-24	Ca-P	

Four different sediment P extractions were carried out in the present study. The first extraction was conducted on untreated sediment and for all collected samples (n=36) in order to assess the P content of each sediment layer. Due to some errors (incomplete homogenization of sediment before subsampling), the procedure was repeated, although the second only included 24 samples, which were carefully chosen (based on the previous mobile P results) to ensure that the entire mobile P range in sediment was adequately represented. The second P extraction was carried out after addition of aluminium sulphate (Al) to 8 samples, corresponding to 8 different concentrations of mobile P in sediment. For each sample, 8 replicates were done, corresponding to different Al dosing ratios. The same was done in the third fractionation, but after Phoslock addition and an extended BD step of 2 hours in total. In both Al and Phoslock additions, the samples were chosen to represent the whole mobile P range in sediments. Likewise, dosing ratios were chosen to both under-dose and over-dose P. A fourth fractionation after Phoslock addition (only for the 4 samples with highest mobile P content) was performed to confirm the results obtained in the first Phoslock fractionation. Moreover, in this last fractionation, the extraction scheme was adapted and an additional BD step of 12 hours in total was performed to assess whether more time would allow Phoslock to bind more P (*Table 2*). Both obtained Fe-P fractions were added together to measure the total P bound to Fe.

Table 2, Adapted P fractionation scheme used for sediment analyses after Phoslock treatment based on (Hupfer et al., 1995) and (Psenner et al., 1988).

Step	Extractant	Time (h)	Fraction	Org-P
1	Double-deionized water (H_2O·P)	2	Labile P	
2*	$NaHCO_3$ 0.1M / Na_2SO_4 0.1M (BD·P)	12	Fe-P	Calculated as the difference Tot-P – Al-P
2	$NaHCO_3$ 0.1M / Na_2SO_4 0.1M (BD·P)	2	Fe-P	
3	NaOH 0.1M (NaOH·P)	16	Al-P	
4	NaOH 0.1M digested with $K_2S_2O_8$ (NaOH·P)		Tot-P	
5	HCl 0.5M (HCl·P)	16-24	Ca-P	

3.5. Dose calculation

Doses of Al and Phoslock were calculated based on mobile P content in the collected sediment samples (*Figure 4*, *Table 24* in Appendix). According to analysed estimates of Mobile P, 8 samples with representative Mobile P concentrations were chosen for the dosing experiments. Dosing ratios were chosen to both under-dose and over-dose mobile P with respect to current dosing recommendations (Rydin et al. (2000), Reitzel et al. (2005), Huser and Pilgrim (2014)). Therefore, ratios for Al (Al:Mobile P) were 0, 5, 10, 25, 50, 75, 100, and 150:1, whereas Phoslock ratios (Phoslock:Mobile P) were 0, 15, 30, 75, 150, 225, 300, and 450:1. From those, Al and Phoslock doses were calculated.

3.6. Dosing experiment

3.6.1. Aluminium experimental set up

The addition of Al sulphate (Alum, $Al_2(SO_4)_3 \cdot 18(H_2O)$) to double deionized water (Al solution) can lead to changes in the solution's acidity which, together with its conductivity due to SO_4^{-2} ions, affects its efficiency to bind mobile P. Therefore, in order to compensate all these effects, the Al solution (~ 4.4g Alum/l) was adjusted to pH 6.7 with NaOH 2M resulting in a final Al concentration of 353 mg/l. Moreover, ionic strength was regulated with NaCl.

Al was added in solution to all untreated sediment sub-samples (~10 mg dry weight) in 15 mL centrifuge tubes according to the required and calculated Al doses. Samples were shaken for 48 hours and then fractionated to determine mobile P, Al-P, Org-P, and Ca-P according to the fractionation procedure mentioned above.

3.6.2. Phoslock experimental set up

The addition of Phoslock (clay) to double deionized water (Phoslock solution) does not lead to changes in the solution's acidity or conductivity, therefore, its efficiency to bind mobile P is not affected and no additional treatments were needed. The Phoslock solution was then prepared by adding the clay to double deionized water until the final concentration of 1000 mg/l was reached. The pH of the solution was approximately 6.8.

The slurry was then diluted to 10 ml and added to each untreated sediment sub-sample (~10 mg dry weight) according to the required and calculated Phoslock doses. Samples were shaken for 48 hours and then fractionated to determine mobile P, Al-P, Org-P, and Ca-P according to the fractionation procedure mentioned above.

4. Results

Results of the performed experiments with sediment sub-samples from both V1 and V2 cores are presented in the below sections. The third core that was sampled (V3) was not included in the laboratory analyses and, therefore, is not presented nor considered in any of the calculations because the core was mostly sand and contained very little sediment.

At the start of the project, some sediment samples were not completely mixed before analysing (first run), giving deviated results. Thus, some analyses were repeated (second run) to improve the results. However, in some cases, estimations from the *first run* are also presented because, even if they are not totally reliable, they can be used as a reference. Correct and definitive results will be referred as *second run*.

Moreover, when analysing sediments after Phoslock treatment (first addition), the laboratory equipment that was being used was damaged, forcing to change the measuring tool. The results obtained were, again, deviated and not representative due to different calibration and analytical method. Therefore, a *second addition* of Phoslock was performed and analysed adequately, this time with fewer sub-samples.

4.1. Basic physical properties of sediments

The water content of the sediments was estimated and results range from 95.4% to 86.4% in core V1, and from 92.8% to 83.4% in core V2 for the second run (sediments well mixed). Results are quite similar to the *first run* and they are all displayed in *Table 16* and *Table 17* in the Appendix.

Density of sediments was estimated with two different methods and detailed results are shown in *Table 18* and *Table 19* in the Appendix. Density, estimated following the mathematical expression based on water content (second run), ranged from 1.010 to 1.069 g/cm^3 for V1 core, and from 1.027 to 1.090 g/cm^3 for V2 core, whereas density estimates after loss on ignition ranged from 1.020 to 1.064 g/cm^3 for V1 core, and from 1.033 to 1.085 g/cm^3 for V2 core. Estimated densities were quite similar, concluding that density can be determined mathematically, giving reasonable good results. All analyses in the present work are based on densities estimated mathematically.

4.2. P content in fresh sediments

4.2.1. Fractionation (first run)

Sediment P content of fresh sediments was initially determined based on the estimated water content and densities from the *first run* for all 36 collected samples (*Figure 3*). The fractionation procedure applied gave values of P content bound to different fractions (mobile P, Al-P, Org-P, Ca-P), which showed the distribution of P in lake sediments. As mentioned before, these results were not representative of the actual distribution of P in sediments, but are still presented for reference. Moreover, in this case, centrifugation of NaOH extracts before analysing Al-P was not performed, resulting in higher Al-P values likely due to suspended organic material in the extracted solution.

As can be observed in the figure, total P content in the V1 core is higher than in core V2. Furthermore, V1 also had higher mobile P content, and it was also greater relative to the other fractions. Fractions Al-P, Org-P, and Ca-P present similar trends and values of P for the whole core in both V1 and V2, although Org-P and Al-P are a bit higher in V1. Detailed results for

measured absorbance and P content (mg/g) are included in the Appendix and presented in *Table 20*, *Table 21*, and *Table 22*.

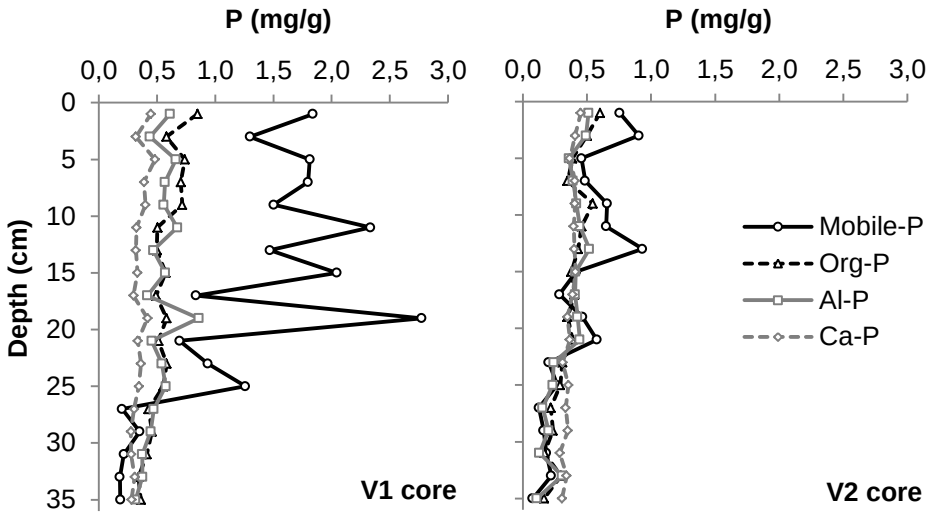

Figure 3, P content (mg/g) in sediments according to depth of sediment (cm) for cores V1 and V2. First run.

4.2.2. Fractionation (second run)

A second fractionation based on the improved water content and densities (second run) was performed, and P contents (mg/g) for each fraction were estimated. In this case, though, not all samples were included in the experiment. 12 samples of each core were chosen, based on the results from the *first run* fractionation and trying to cover the whole mobile P range of the sediments. Only mobile P was analysed.

As can be observed in *Figure 4*, total P content in V1 core is higher than in core V2. Results follow the same trend observed in *Figure 3*, although improved values of mobile P in V1 are lower than results from the first fractionation, whereas V2 values are similar. Detailed results are included in the Appendix (*Table 23* and *Table 24*).

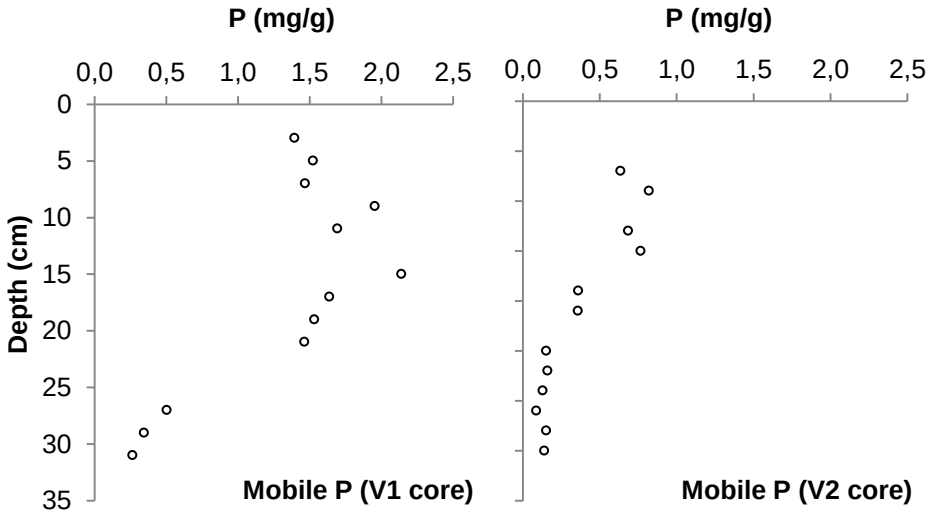

Figure 4, P content (mg/g) in sediments according to depth of sediment (cm) for cores V1 and V2 (second run).

4.3. P content in sediments after treatment

4.3.1. Selection of samples

According to the fractionation results (second run), a set of 8 samples with different mobile P content was selected, trying to cover the entire P range of the sediments. These samples were later treated with Al and Phoslock (*Table 3*).

Table 3, Selection of samples to dose. Sample number, sample name, sample layer, measured mobile P mass (in g/m^2/cm) and concentrations (in mg/g) are indicated.

Sample	***Name*** *(core_ID)*	***Layer*** *(cm)*	***Mobile P*** *(g/m^2/cm)*	***Mobile P*** *(mg/g)*
1	V1_8	14-16	2.06	2.14
2	V1_11	20-22	1.63	1.46
3	V1_5	8-10	1.55	1.96
4	V1_3	4-6	1.20	1.52
5	V1_2	2-4	1.02	1.40
6	V2_7	12-14	0.79	0.69
7	V2_10	18-20	0.43	0.36
8	V2_15	28-30	0.21	0.13

4.3.2. Aluminium

The 8 selected samples (*Table 3*) were treated with Al (see doses in Appendix, *Table 25*) and later analysed (via fractionation) to determine P content distribution in the sediment after treatment. As mentioned before, eight different Al:Mobile P ratios (0, 5, 10, 25, 50, 75, 100, 150) were applied to both under-dose and over-dose P.

Detailed results for measured absorbance and P content (mg/g) after Al treatment are included in the Appendix in *Table 26*, *Table 27*, *Table 28*, *Table 29*, *Table 30*, and *Table 31*, where both Al-P and Org-P distributions (with or without centrifugation of NaOH extracts before analysing) are also presented to allow their comparison. As an example, *Figure 5* shows a comparison of Al-P and Org-P results for sample 1 (see *Table 3*) with (yes) and without (no) centrifugation of NaOH extracts. As can be observed, centrifugation reduced the presence of organic matter when analysing Al-P, lowering its P content, while increasing P content of Org-P.

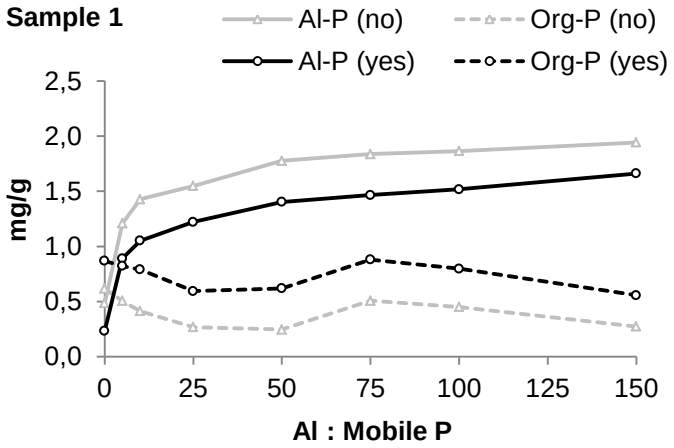

Figure 5, Al-P and Org-P content (mg/g) in sample 1 after Al treatment. Comparison of centrifugation effect in Al-P and Org-P fractions. "No" refers to measurement without centrifugation and "yes" with centrifugation.

Results from fractionations that include the centrifugation of NaOH extracts before analysis of Al-P were considered more realistic due to the reduction of organic matter in the Al-P fraction and lower interference with light absorption in the spectrophotometer, leading to more accurate values of Al-P and, therefore, more accurate values of Org-P too.

Measured mobile P in sediment (ratio 0, *Table 26*) was generally lower than the reference mobile P (*Table 3*) used to choose the samples to dose, even though trends were the same (*Figure 6*). Moreover, Org-P content decreased when mobile P decreased across the different samples. Furthermore, in samples with higher mobile P (samples 1 to 5), Org-P also decreased with increasing Al doses, whereas, in samples 6 to 8, it remained more or less constant. On the contrary, Ca-P shows to be constant across all samples and not affected by Al addition.

Sediments with higher P concentrations required lower Al doses than sediments with lower P concentrations to reach 80-90% of mobile P reduction (*Table 4*). Samples 7 and 8 did not even reach a 90% of P removal with highest dose (150:1 ratio). In terms of dosage efficiency, Al:Mobile P ratios giving maximum P reduction with the lowest dose possible (from now on referred as optimal) varied depending on mobile P content of the sediment, as can be observed in *Figure 6*. For samples 1 to 6, 75 was the optimal Phoslock:Mobile P ratio, whereas for samples 7 and 8 the ratio was 25 (due to their low P content).

Table 4, Reduction of mobile P with increasing Al doses (Al:Mobile P ratios of 0, 5, 10, 25, 50, 75, 100, 150). Bold values indicate optimal P removal.

Sample	***Layer***	***Mobile P***	***Ratios / Mobile P reduction (%)***							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	1.55	0%	44%	55%	73%	84%	**90%**	92%	95%
2	20-22	1.20	0%	44%	53%	73%	82%	**88%**	91%	94%
3	8-10	1.66	0%	52%	52%	73%	82%	**88%**	91%	95%
4	4-6	1.59	0%	49%	55%	66%	83%	**87%**	88%	93%
5	2-4	1.53	0%	35%	44%	61%	73%	**83%**	86%	89%
6	12-14	0.64	0%	37%	47%	62%	72%	**80%**	84%	86%
7	18-20	0.33	0%	35%	43%	**63%**	69%	76%	79%	85%
8	28-30	0.12	0%	-2%	23%	**52%**	59%	70%	71%	79%

Mobile P was definitely reduced with increasing doses of Al. Approximately all mobile P was adsorbed by Al, observable by the mobile P loss and the increase of the Al-P fraction (*Table 5*).

Table 5, Maximum mobile P loss (mg/g), maximum increase of Al-P fraction (mg/g), and remaining mobile P (mg/g, difference).

Sample	***Layer*** *(cm)*	***Maximum mobile P loss***	***Maximum Al-P increase***	***Remaining mobile P***
1	14-16	1.46	1.43	0.04
2	20-22	1.14	0.99	0.15
3	8-10	1.58	1.27	0.31
4	4-6	1.48	1.23	0.25
5	2-4	1.36	1.31	0.06
6	12-14	0.56	0.49	0.07
7	18-20	0.28	0.26	0.02
8	28-30	0.09	0.06	0.03

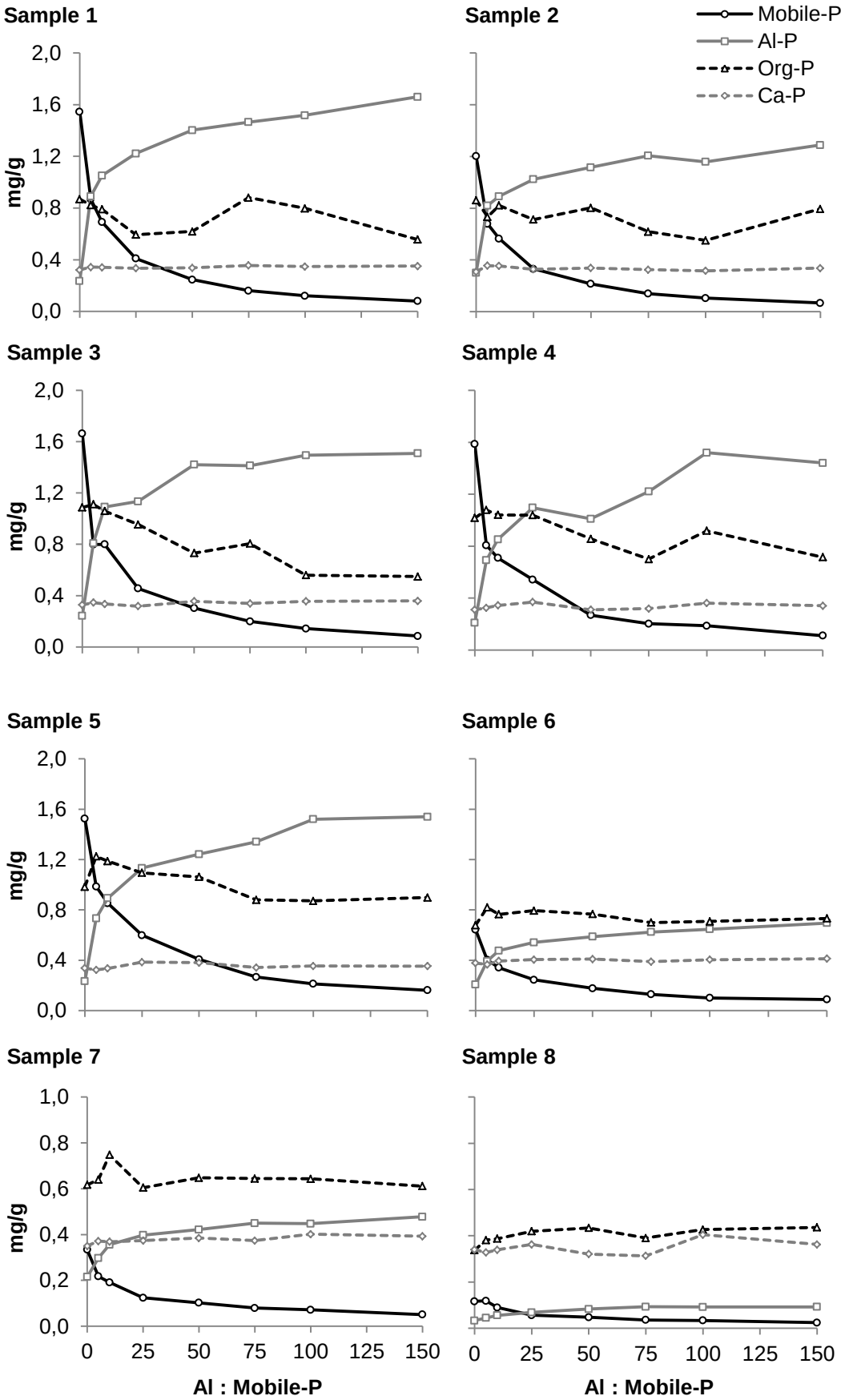

Figure 6, P content (mg/g) distribution in sediments for selected samples (1-8) after Al treatment. Mobile P fraction includes both labile and Fe-P fractions. Al-P and Org-P fractions were estimated after centrifugation of NaOH extract.

Detailed results of binding efficiency ratios (Al:Al-P) and ratios between Al added and mobile P lost (Al:Mobile P_L) are presented in the Appendix (*Table 32* and *Table 33*). For the 6 samples with the optimal ratio (Al:Mobile P) of 75:1, Al:Al-P binding ratios ranged from 78 to 110 (by weight). Ratios between Al added and mobile P lost (Al:Mobile P_L), for these same samples and ratio, ranged from 83 to 116 (by weight).

In addition, theoretical binding ratios, calculated as the ratio of Al to available P for adsorption (mobile P measured at dose Al:Mobile P ratio of 0), for samples 1-6 with optimal Al:Mobile P ratio of 75, ranged from 9.62 to 14.58 mg P/g Al, whereas actual binding ratios (P lost and bound to Al) for those same samples ranged from 8.62 to 12.12 mg P/g Al. Such close results show a great real P removal capacity of Al. *Table 6* shows a comparison of these ratios for the Al:Mobile P dose ratio of 75 (see *Table 34* and *Table 35* in the Appendix for complete results). The comparison between theoretical and actual/real binding ratios showed that higher Al doses brought both ratios closer together, whereas the largest difference was observed at low Al doses.

Table 6, Theoretical and actual binding ratios (mg P/g Al) for the 8 treated samples at the Al:Mobile P dose ratio of 75. Mobile P values used to calculate the theoretical ratio correspond to measured mobile P at Al dose ratio of 0.

Sample	***Layer (cm)***	***Mobile P (mg/g)***	***Al dose (mg/g)***	***Theoretical (mg P/g Al)***	***Actual (mg P/g Al)***
1	14-16	1.55	160.58	9.62	8.62
2	20-22	1.20	109.74	10.97	9.70
3	8-10	1.66	146.63	11.35	10.00
4	4-6	1.59	114.25	13.88	12.12
5	2-4	1.53	104.64	14.58	12.03
6	12-14	0.64	51.40	12.52	10.01
7	18-20	0.33	27.09	12.32	9.38
8	28-30	0.12	9.80	11.83	8.22

In current lake restoration projects, doses are typically chosen so that internal P loading rates meet a specific goal, therefore, low P concentrations are not usually targeted as they do not represent high risks for eutrophication. Potential internal P release rates (Li, $mg/m^2/d$) were also estimated in order to see the minimum release rate achievable with Al treatment (Huser and Pilgrim, 2014), which were calculated based on the mass of mobile P ($g/m^2/cm$) still remaining in the sediment layer. For Al treatment, Li ranged from 2.23 to 0.15 $mg/m^2/d$ at the cost-effective Al:Mobile P ratio of 75, whereas values were close to 0 at the highest ratio (150:1), meaning that potential P cycling was almost eliminated (92-98% reduction). Detailed potential internal P release rates for all samples at each ratio are included in the Appendix (*Table 36* and *Table 37*).

4.3.3. Phoslock (first addition)

The 8 selected samples (*Table 3*) were treated with Phoslock (see doses in Appendix, *Table 38*) and later analysed (via fractionation) to determine P content in the sediment after treatment. As mentioned before, eight different Phoslock:Mobile P ratios (0, 15, 30, 75, 150, 225, 300, 450) were applied to both under-dose and over-dose P.

In the middle of this experiment, the analytical instrument that was being used broke, preventing the measurement of the NaOH extracts after centrifugation (Al-P fraction). Consequently, obtained estimations for Al-P and Org-P (without centrifugation) were somewhat deviated, giving higher and lower values, respectively. Nevertheless, results are validated by the process

applied (Murphy and Riley (1962), Psenner et al. (1988), Hupfer et al. (1995)), but they cannot be compared with the Al treatment results (with centrifugation) presented in section 4.3.2. Moreover, due to the damaged machine, fractions Org-P and Ca-P from samples 1 to 4 and all fractions from samples 5 to 8 were measured with a different instrument than labile P, Fe-P and Al-P fractions from samples 1-4. Therefore, global results from this fractionation gave an idea of P behaviour and distribution across fractions, but estimations between fractions were not comparable. In addition, the analytical instrument seemed to give strange measurements; hence, results presented below are neither reliable nor representative.

Detailed results for measured absorbance and P content (mg/g) after Phoslock treatment are included in the Appendix in *Table 39*, *Table 40*, *Table 41*, and *Table 42*.

As can be observed in *Figure 7*, measured mobile P in sediment (ratio 0, *Table 39*) was generally lower than the reference mobile P (*Table 3*) used to choose the samples to dose. It can also be seen that Al-P and Org-P content remained more or less constant across all samples and were not affected by Phoslock addition, whereas Ca-P increased with Phoslock addition. Measurements for samples 7 and 8 were very variable, giving very strange results that were not always taken into consideration. Reasons for this could be the low P concentrations and the errors of the analytical instrument.

Phoslock:Mobile P ratios giving maximum P reduction with the lowest dose possible also varied depending on mobile P content of the sediment (*Figure 7*). For the majority of the samples (2 to 6), the Phoslock:Mobile P ratio giving maximum P removal was 225, whereas for samples 1 and 8 the ratio was 150, and for sample 7 the ratio was 75. In all cases, such ratios were equivalent to a removal of P between 30-40%, approximately. It can also be observed, in some samples, how P removal decreased with increasing doses after reaching the point of maximum P adsorption (see *Table 7* and *Figure 7*). This same trend could be observed, in some samples, at the lowest doses.

Table 7, Reduction of mobile P with increasing Phoslock doses (Phoslock:Mobile P ratios of 0, 15, 30, 75, 150, 225, 300, 450). Results are not totally reliable due to the analytical instruments used. Bold indicates maximum reduction of P.

Sample	***Layer***	***Mobile P***	***Ratios / Mobile P reduction (%)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	1.63	0%	2%	4%	14%	**34%**	32%	36%	29%
2	20-22	1.16	0%	1%	-1%	8%	25%	**36%**	38%	29%
3	8-10	1.56	0%	1%	1%	8%	22%	**31%**	25%	23%
4	4-6	1.59	0%	0%	3%	9%	17%	**27%**	27%	29%
5	2-4	1.56	0%	4%	1%	12%	-4%	**38%**	34%	43%
6	12-14	0.61	0%	5%	6%	-11%	16%	**33%**	32%	40%
7	18-20	0.43	0%	-30%	31%	**36%**	4%	-7%	41%	58%
8	28-30	0.09	0%	-46%	-76%	-12%	**82%**	-171%	-142%	99%

Mobile P was reduced with increasing doses of the binding agent, but not drastically. Approximately, less than half of the mobile P was adsorbed by Phoslock, which was observable by the increase of the Ca-P fraction, meaning that mobile P was bound to Phoslock and extracted in the Ca-P fraction. However, Ca-P increase was higher than mobile P lost in some samples (see *Table 8*). This phenomenon could be due to the different instruments with which mobile P and Ca-P were measured, but also due to P coming from some other P forms than mobile P, maybe even from the residual P fraction, which was not analysed in these experiments.

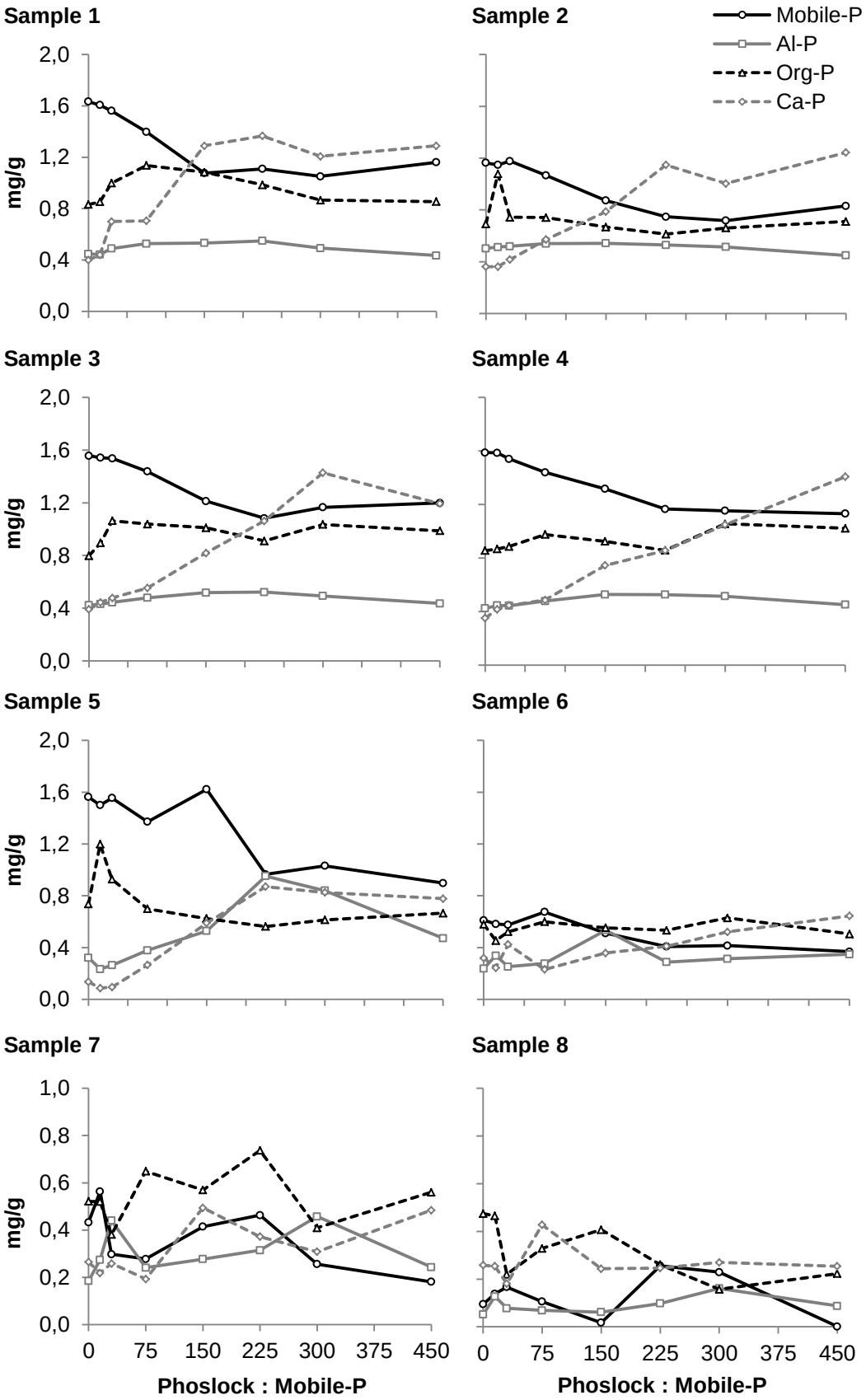

Figure 7, P content (mg/g) and distribution in sediments for selected samples after Phoslock treatment. Mobile P fraction includes labile and Fe-P fractions. Mobile P and Al-P for samples 1 to 4 were analysed with a different machine than Org-P and Ca-P fractions and all fractions for samples 5 to 8. Al-P and Org-P fractions were estimated without centrifugation of NaOH extract.

Table 8, Maximum reduction of mobile P (mobile P loss, mg/g), maximum increase of Ca-P fraction (mg/g), and remaining mobile P (mg/g, difference).

Sample	***Layer*** *(cm)*	***Maximum mobile P loss***	***Maximum Ca-P increase***	***Remaining mobile P***
1	14-16	0.58	0.97	-0.38
2	20-22	0.46	0.88	-0.42
3	8-10	0.48	1.04	-0.56
4	4-6	0.46	1.06	-0.60
5	2-4	0.72	0.78	-0.06
6	12-14	0.31	0.41	-0.11
7	18-20	0.38	0.30	0.08
8	28-30	0.26	0.25	0.01

Detailed results of binding efficiency ratios (Phoslock:Al-P and Phoslock:Ca-P) and ratios between Phoslock added and mobile P lost (Phoslock:Mobile P_L) are presented in the Appendix (*Table 43*, *Table 44*, and *Table 45*). For the 5 samples with the ratio (Phoslock:Mobile P) of 225:1, Phoslock:Ca-P binding ratios ranged from 418 to 538 (by weight). Ratios between Phoslock added and mobile P lost (Phoslock:Mobile P_L), for these same samples and ratio, ranged from 525 to 925 (by weight).

In addition, theoretical binding ratios, calculated as the ratio of Phoslock to available P for adsorption (mobile P measured at dose Phoslock:Mobile P ratio of 0), for samples 2-6 with Phoslock:Mobile P ratio of 225, ranged from 3.53 to 4.98 mg P/g Phoslock, whereas actual binding ratios (P lost and bound to Phoslock) for those same samples ranged from 1.08 to 1.90 mg P/g Phoslock. Such different ratios show the real low P removal capacity of Phoslock. *Table 9* shows a comparison of these ratios for the Phoslock:Mobile P dose ratio of 225 (see *Table 46* and *Table 47* in the Appendix for complete results). The comparison between theoretical and actual/real binding ratios showed that higher Phoslock doses brought both ratios closer together, whereas the largest difference was observed at low Phoslock doses.

Table 9, Theoretical and actual binding ratios (mg P/g Phoslock) for the 8 treated samples at the Phoslock:Mobile P dose ratio of 225. Mobile P values used to calculate the theoretical ratio correspond to measured mobile P at Phoslock dose ratio of 0.

Sample	***Layer (cm)***	***Mobile P (mg/g)***	***Phoslock dose (mg/g)***	***Theoretical (mg P/g Phoslock)***	***Actual (mg P/g Phoslock)***
1	14-16	1.63	481.73	3.39	1.09
2	20-22	1.16	329.21	3.53	1.26
3	8-10	1.56	439.90	3.54	1.08
4	4-6	1.59	342.75	4.63	1.23
5	2-4	1.56	313.91	4.98	1.90
6	12-14	0.61	154.19	3.96	1.31
7	18-20	0.43	81.27	5.32	-0.38
8	28-30	0.09	29.39	3.22	-5.50

Potential internal P release rates (Li, $mg/m^2/d$) were also estimated in order to see the minimum release rate achievable with Phoslock treatment (Huser and Pilgrim, 2014), which were calculated based on the mass of mobile P ($g/m^2/cm$) still remaining in the sediment layer. For Phoslock treatment, Li ranged from 15.43 to 5.49 $mg/m^2/d$ at the Phoslock:Mobile P ratio of 225, meaning that the maximum achievable reduction of potential P cycling (28-40%) might not

reach management goals because a large amount of P still remained in the sediment after treatment (values should be close to 0). Detailed potential internal P release rates for all samples at each ratio are included in the Appendix (*Table 48* and *Table 49*).

Even though the above results gave an idea of P fractionation in sediments after Phoslock treatment, they will no longer be discussed due to all the issues already mentioned.

4.3.4. Phoslock (second addition)

Due to the problems and the associated issues with the results obtained in the first addition of Phoslock (section 4.3.3), a second fractionation after Phoslock addition (only for samples 1 to 4, *Table 3*) was performed to compare it with the results displayed in *Figure 7*.

The samples were treated with Phoslock (see doses in Appendix, *Table 50*) and later analysed (via fractionation) to determine P content distribution in the sediment after treatment. As mentioned before, eight different Phoslock:Mobile P ratios (0, 15, 30, 75, 150, 225, 300, 450) were applied to both under-dose and over-dose P.

Detailed results for absorbance and P content (mg/g) after Phoslock treatment are included in the Appendix in *Table 51*, *Table 52*, *Table 53*, *Table 54*, *Table 55*, and *Table 56*, where both Al-P and Org-P distributions (with or without centrifugation of NaOH extracts before analysing) are also presented to allow their comparison. As an example, *Figure 8* shows a comparison of Al-P and Org-P results for sample 1 (*Table 3*) with (yes) and without (no) centrifugation of NaOH extracts. As can be observed, centrifugation reduced the presence of organic matter when analysing Al-P, lowering its P content, while increasing P content of Org-P.

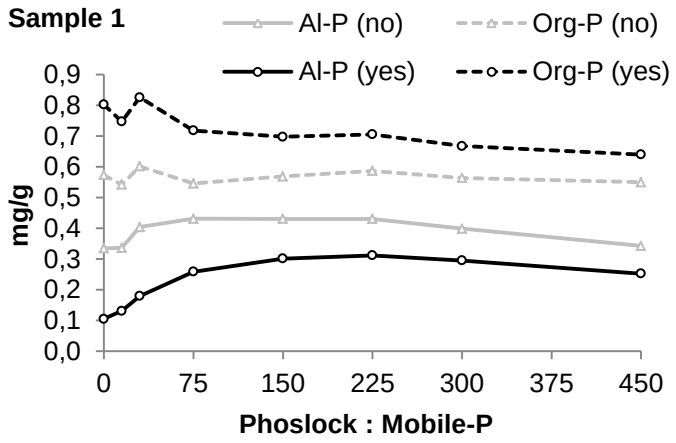

Figure 8, Al-P and Org-P content (mg/g) in sample 1 after Phoslock treatment. Comparison of centrifugation effect in Al-P and Org-P fractions. "No" refers to measurement without centrifugation and "yes" with centrifugation.

Results from fractionations that include the centrifugation of NaOH extracts before analysis of Al-P were considered more realistic due to the reduction of organic matter in the Al-P fraction and the lower interference with light absorption in the spectrophotometer, leading to more accurate values of Al-P and, therefore, more accurate values of Org-P too.

As can be observed in *Figure 9*, measured mobile P in sediment (ratio 0, *Table 51*) was generally lower than the reference mobile P (*Table 3*) used to choose the samples to dose. It can also be seen that Al-P and Org-P content remained more or less constant across all samples and was not affected by Phoslock addition, whereas Ca-P increased with Phoslock addition (until reaching some stabilization).

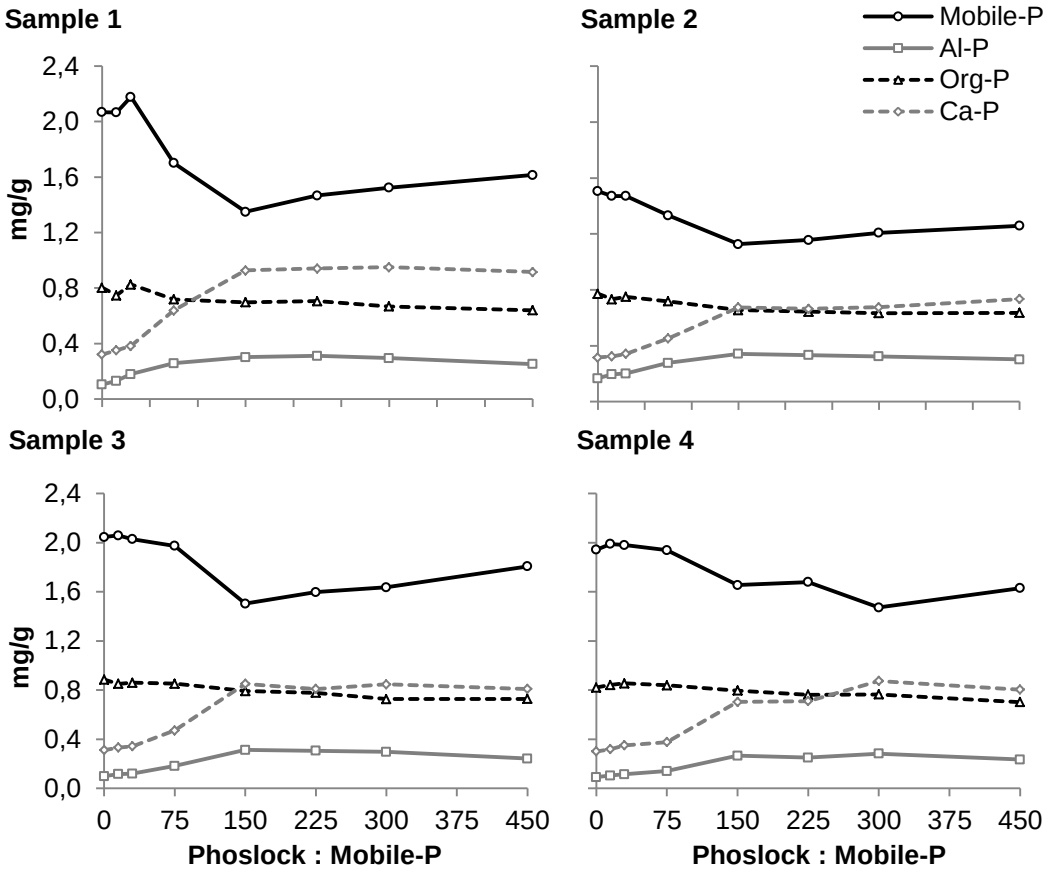

Figure 9, P content (mg/g) and distribution in sediments for selected samples after Phoslock treatment. Mobile P fraction includes both labile and Fe-P fractions. Al-P and Org-P fractions were estimated after centrifugation of NaOH extract.

Phoslock:Mobile P ratios giving maximum P reduction with the lowest dose possible also varied depending on mobile P content of the sediment, as it is displayed in *Figure 9*. For samples 1-3, 150 was the Phoslock:Mobile P ratio giving maximum P removal, whereas for sample 4 the ratio was 300. Such ratios all gave P reduction efficiencies between 25-35% (see *Table 10*). According to results, sediments with higher P concentrations require lower Phoslock doses than sediments with lower P concentrations to reach a similar mobile P reduction. Even though there is some variability, results also showed how P removal decreased with increasing doses after reaching the point of maximum P adsorption (see *Table 10* and *Figure 9*). This same trend could be observed, in some samples, at the lowest doses.

Table 10, Reduction of mobile P with increasing Phoslock doses (Phoslock:Mobile P ratios of 0, 15, 30, 75, 150, 225, 300, 450). Bold indicates maximum reduction of mobile P

Sample	***Layer***	***Mobile P***	***Ratios / Mobile P reduction (%)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.07	0%	0%	-5%	18%	**35%**	29%	26%	22%
2	20-22	1.51	0%	2%	2%	11%	**25%**	23%	20%	16%
3	8-10	2.05	0%	-1%	1%	4%	**27%**	22%	20%	12%
4	4-6	1.94	0%	-2%	-2%	0%	15%	14%	**24%**	16%

Regarding binding efficiency of Phoslock, mobile P was reduced with increasing doses of the binding agent, but not drastically. Approximately, less than half of the mobile P was adsorbed or fixed by Phoslock, which was observable by the increase of the Ca-P fraction, meaning that

mobile P was bound to Phoslock and then released from the La ions under low pH conditions. However, Ca-P increase was higher than mobile P lost in some samples (see *Table 11*), meaning that P present in the Ca-P fraction was also coming from some other P forms, maybe even from the residual P fraction, which was not analysed in these experiments, but, in any case, to a much lesser extent than in the first Phoslock addition (section 4.3.3).

Table 11, Maximum reduction of mobile P (mobile P loss, mg/g), maximum increase of Ca-P fraction (mg/g), and remaining mobile P (mg/g, difference).

Sample	***Layer*** *(cm)*	***Maximum mobile P loss***	***Maximum Ca-P increase***	***Remaining mobile P***
1	14-16	0.83	0.63	0.20
2	20-22	0.38	0.42	-0.04
3	8-10	0.56	0.54	0.02
4	4-6	0.52	0.57	-0.06

Detailed results of binding efficiency ratios (Phoslock:Al-P and Phoslock:Ca-P) and ratios between Phoslock added and mobile P lost (Phoslock:Mobile P_L) are presented in the Appendix (*Table 57*, *Table 58*, and *Table 59*). For the 3 samples with the ratio (Phoslock:Mobile P) of 150:1, Phoslock:Ca-P binding ratios ranged from 325 to 346 (by weight). Ratios between Phoslock added and mobile P lost (Phoslock:Mobile P_L), for these same samples and ratio, ranged from 447 to 579 (by weight).

Theoretical binding ratios, calculated as the ratio of Phoslock to available P for adsorption (mobile P measured at dose Phoslock:Mobile P ratio of 0), for samples 1-3 with Phoslock:Mobile P ratio of 150, ranged from 6.44 to 6.98 mg P/g Phoslock, whereas actual binding ratios (P lost and bound to Phoslock) for those same samples ranged from 1.73 to 2.24 mg P/g Phoslock. Such different ratios show the real low P removal capacity of Phoslock. *Table 12* shows a comparison of these ratios for the Phoslock:Mobile P dose ratio of 150 (see *Table 60* and *Table 61* in the Appendix for complete results). The comparison between theoretical and actual/real binding ratios showed that higher Phoslock doses brought both ratios closer together, whereas the largest difference was observed at low Phoslock doses.

Table 12, Theoretical and actual binding ratios (mg P/g Phoslock) for the 4 treated samples at the Phoslock:Mobile P dose ratio of 150. Mobile P values used to calculate the theoretical ratio correspond to measured mobile P at Phoslock dose ratio of 0.

Sample	***Layer (cm)***	***Mobile P (mg/g)***	***Phoslock dose (mg/g)***	***Theoretical (mg P/g Phoslock)***	***Actual (mg P/g Phoslock)***
1	14-16	2.07	321.16	6.44	2.24
2	20-22	1.51	219.48	6.86	1.73
3	8-10	2.05	293.27	6.98	1.85
4	4-6	1.94	228.50	8.50	1.26

Potential internal P release rates (Li, $mg/m^2/d$) were also estimated in order to see the minimum release rate achievable with Phoslock treatment (Huser and Pilgrim, 2014), which were calculated based on the mass of mobile P ($g/m^2/cm$) still remaining in the sediment layer. For Phoslock treatment, Li ranged from 17.32 to 18.96 $mg/m^2/d$ at the Phoslock:Mobile P ratio of 150, meaning that the maximum achievable reduction of potential P cycling (15-36%) might not reach management goals because a large amount of P still remained in the sediment after

treatment (values should be close to 0). Detailed potential internal P release rates for all samples at each ratio are included in the Appendix (*Table 62* and *Table 63*).

5. Relative costs of treatment

To compare relative treatment costs, prices and P removal capacity for both products were considered. Phoslock price of 2750 €/ton was found in Mackay et al. (2014), whereas Al price (1.6 $US/gallon) was found in page 22 of Barr Engineering Company (2012) and was converted to 0.66 $US/kg Alum ($Al_2(SO_4)_3 \cdot 14(H_2O)$) with Alum properties (5.4 lbs Alum/gallon) found in C&S Chemicals, Inc. (2000). From that, the price of 6432 €/ton Al was estimated.

In order to compare costs of an equivalent P reduction (in %), treatment costs for a P removal of 35% in Växjö Lake sediments were estimated, since Phoslock can only reach such maximum P reduction. Under these conditions, costs per ton of sediment and per kg of P removed were calculated based on the required doses and P removed (mass) for each treatment. Based on the study results, Phoslock treatment would cost 883 €/ton of sediment or 1227 €/kg of P removed, whereas Al treatment would cost 45 €/ton of sediment or 83 €/kg of P removed. *Table 13* shows the data used to estimate such costs.

Table 13, Costs comparison for Al and Phoslock treatments (35% P removal) in Växjö Lake.

Concept	***Al***	***Phoslock***
Price per ton of product	6432 €/ton	2750 €/ton
Doses required for 35% P removal	6.98 mg/g sediment	321.16 mg/g sediment
Cost per ton of sediment	***45 €/ton sediment***	***883 €/ton sediment***
P removed with treatment	0.54 mg P/g sediment	0.72 mg P/g sediment
Cost per kg P removed	***83 €/kg P removed***	***1227 €/kg P removed***

Besides the costs of the product to apply, extra costs might need to be considered in each treatment. For example, if an alkalinity buffer or the addition of other products were to be required before adding Al to sediments to ensure an environmentally safe treatment, such costs should be computed as well. Likewise, longevity of the treatment, aging of the added product, and dosing repetition (if required) would need to be considered when computing total costs of the treatment (Lewandowski et al. (2003b), de Vicente et al. (2008), Huser et al. (2015)).

In any case, and as presented above, Phoslock treatment appears to be over an order of magnitude more expensive than Al treatment, even with the extra costs that the latter might require.

6. Discussion

Dosing relationships developed and binding ratios estimated in this thesis showed how Al treatment was more effective than Phoslock treatment in terms of adsorption of available P in sediments. Al application appeared to bind almost all mobile P, whereas Phoslock was only able to remove less than half of the mobile P content in sediments.

6.1. Al treatment

The study showed that mobile P was definitely reduced with the addition of Al, meaning that adsorption capacity of sediments increased with such treatment, as also stated by Rydin and Welch (1999), Lewandowski et al. (2003b), and Reitzel et al. (2005), among others. After Al application, mobile P reductions greater than 90% were observed in the samples with highest mobile P (1.20-1.66 mg/g) and at Al:Mobile P ratios of 75 (90%), 100 (91-92%) and 150 (93-95%). Similar P reductions were found by Pilgrim et al. (2007), who also stated that P release rates were linearly related to mobile P in sediments (Huser and Pilgrim, 2014).

In terms of Al:Mobile P dosages, results showed that required ratios to remove similar amounts of P (80%, for example) across different samples were variable and decreased with increasing mobile P, which agrees with James (2011). Al:Mobile P ratios of 75 (for samples 1-6, 1.55-0.64 mg mobile P/g sediment) and 25 (for samples 7 and 8, 0.33-0.12 mg mobile P/g sediment) were the most cost-effective in these experiments, reaching great removal efficiencies (90-80% and 52-63%, respectively) with relatively low Al doses (51.40-160.58 mg/g and 3.27-9.03 mg/g, respectively). In comparison, samples 1-6 would need much larger Al doses (102.79-321.16 mg/g) to reach 86-95% of P removal, as well as samples 7 and 8, which would need Al doses between 19.60-54.18 mg/g to reach 79-85% of P removal.

Estimated internal P loading rates after treatment showed that management goals are likely to be met since almost no P content remained in the sediment active layer. Potential cycling P rates ranged between 2.23 and 0.15 mg/m^2/d at the cost-effective Al:Mobile P ratio of 75, and values were close to 0 at the highest Al dose (150:1), which is what is expected (Huser and Pilgrim, 2014). Therefore, P release rates were reduced by more than a 90%, as also stated by Reitzel et al. (2005).

Furthermore, it was also observed that adsorbed mobile P was converted to Al-P, since mobile P loss was almost equivalent to Al-P increase, agreeing with what Rydin and Welch (1999), Hansen et al. (2003), and Reitzel et al. (2005) reported. In addition, Org-P also showed a small decrease with increasing Al doses in some samples, whereas Ca-P remained constant across all samples and not affected by Al addition.

Theoretical and actual/real binding ratios grew closer with increasing Al doses. For samples 1-6 at the 75 dose ratio, theoretical binding ratios ranged from 9.62 to 14.58 mg P/g Al, whereas real ratios ranged from 8.62 to 12.12 mg P/g Al, meaning that Al showed a great P removal capacity.

In summary, the Al treatment applied to Växjö Lake sediments gave results that were very similar to what was previously reported and no discrepancies were found.

6.2. Phoslock treatment

This study showed that Phoslock did reduce mobile sediment P content, but only achieved P removal efficiencies of 24-35%. As with Al, sediments with higher P concentrations required lower Phoslock doses than sediments with less P content to reach similar mobile P reductions

(in %). The Phoslock:Mobile P ratio of 150 was clearly the most effective for samples 1-3, in terms of maximum P removal (25-35%) and relatively low Phoslock dose required (219.48-321.16 mg/g). On the other hand, sample 4 required a Phoslock:Mobile P ratio of 300 (456.99 mg/g) to reach a P removal of 24%. Related to this, estimated internal P loading rates after treatment (Huser and Pilgrim, 2014) showed that management goals might not be achieved (depending on the internal loading reduction goal) since large amounts of P still remained in the sediment active layer. Potential cycling P rates after treatment ranged between 17.32 and 18.96 $mg/m^2/d$ at the Phoslock:Mobile P ratio of 150.

Theoretical and actual/real binding ratios grew closer with increasing Phoslock doses. For samples 1-3 at the 150 dose ratio, theoretical binding ratios ranged from 6.44 to 6.98 mg P/g Phoslock, whereas real ratios ranged from 1.73 to 2.24 mg P/g Phoslock, meaning that Phoslock did not show a great P removal capacity. Such results are quite far from the binding ratios achieved by Reitzel et al. (2013) in lake water. Moreover, results also showed that P removal decreased with increasing doses after reaching the point of maximum P adsorption, agreeing with Reitzel et al. (2013), who found maximum binding ratios at the 11:1 dose, instead of the recommended ~100:1 dose.

After Phoslock treatment, mobile P (mainly Fe-P) loss was accompanied with an increase of the Ca-P fraction, meaning that mobile P was bound to Phoslock and extracted in the Ca-P fraction, as Meis et al. (2012), Meis et al. (2013) and Reitzel et al. (2013) also observed. However, in some samples, results also showed a Ca-P increase that was greater than mobile P loss, indicating that P bound by Phoslock may have come from other P forms, maybe even from the residual P fraction, which was not analysed in the experiments. Furthermore, even though Al-P and Org-P content remained more or less stable across all samples and they did not show a significant effect with Phoslock addition, their small variability may be related to the overall P fractionation process. Such a hypothesis could be in line with Reitzel et al. (2013), who reported that Fe and Al are also present in the Phoslock matrix, confirming the existence of binding sites other than La. In any case, it is clear that more work needs to be done to investigate such phenomenon.

Even though results from the first Phoslock addition cannot be trusted, they shared the same trend with the second addition in that P removal decreased with increasing doses, after reaching the point of maximum P adsorption. This phenomenon was also observed by Reitzel et al. (2013), who suggested that relative adsorption of mobile P to Phoslock declined with increasing dosages. It was also observed that, in both fractionations, this same phenomenon occurred at some of the low doses, as if mobile P content increased momentarily, but this may have been due to simple analytical and subsampling errors or variability that are inherent in these types of experiments.

In summary, the Phoslock treatment applied to Växjö Lake sediments gave results that seemed to be in line with Meis et al. (2012) and Meis et al. (2013) in that, with the exception of Ca-P, other sediment P fractions did not change significantly after Phoslock application, hence, did not remove mobile P efficiently, which was the main goal. Such results did not agree with other Phoslock reports (Douglas et al. (2000), Afsar and Groves (2009), Finsterle (2014), Zamparas et al. (2015)), who stated that Phoslock treatment could achieve P removal efficiencies greater than 95%. However, these reductions were only achieved in water solution (or natural waters). It is possible that more time between Phoslock application and the start of the analyses might have allowed for more binding, but both Finsterle (2014) and Zamparas et al. (2015) showed that most binding generally occurs in the first 2 hours. However, even with an additional 12 hours step in the fractionation scheme, no additional binding was observed. Therefore, it seems reasonable to assume that Phoslock can become a good strategy to effectively remove P in solution and fix it in the sediments to avoid future P cycling, but it is not good enough to

inactivate potentially available P already bound to sediments unless it is used in conjunction with other materials.

6.3. Comparison Al versus Phoslock

In order to give a better overview of both Al and Phoslock products, their limitations, their advantages, and other treatment characteristics, a comparison scheme is presented in *Table 14*.

Table 14, Comparison of characteristics for Al and Phoslock. Effectiveness, main limiting factors, toxicity, doses and costs are indicated. Provided information was collected from reviewed literature and results from this thesis are indicated to allow a comparison (a, b, c, d indexes).

	Al	***Phoslock***
Removal efficiency	~ 90% (P in solution and sediments) [a]	~ 90% (P in solution) [c]
Effective pH	6-9	4.5 – 8.5
Effective alkalinity	High is better	Low is better but is not a relevant factor
Sediment properties after treatment	High sedimentation rate, can be released after resuspension, no improvement in sediment consolidation	Improved consolidation and compaction potential, increased erosion threshold
Improvement of light climate	Very good	Worse than Al
Toxicity	Al^{+3} and $Al(OH)_2^+$ if pH<5.5 or pH>9.5 and low alkalinity	La^{+3} if high alkalinity and low and high pH
Chemical interferences	Sulphates, fluorides, phosphate, organo-Al complexes, organic matter, silicates	Ca^{+2}, humic compounds, dissolved organic carbon
Other factors reducing removal efficiency	Lake morphology, aging, mobility, resuspension, benthic disturbances, focusing	La translocation due to resuspension by wind or bioturbation
Treatment longevity	<1 – 40+ years	Not enough information
Dose range (ratio to mobile P)	10 – 100 [b]	100 (or higher) [d]
Cost	83 €/kg P removed	1227 €/kg P removed

[a] *Result of this thesis: ~ 90%*
[b] *Result of this thesis: 75*
[c] *Result of this thesis: 25 – 35% (P in sediments)*
[d] *Result of this thesis: 150 (or higher)*

Only considering all the information gathered from reviewed literature, it is clear that Al has been used for a longer period and is better studied than Phoslock, thereby its application seems safer as long as potential weaknesses are controlled.

Reported P removal efficiency is similar for both products (Douglas et al. (2000), Afsar and Groves (2009), Finsterle (2014), Zamparas et al. (2015), Pilgrim et al. (2007)). However, Al treatment is effective at binding P in all forms (in solution but also already in sediments), whereas Phoslock seems to be more effective at binding P in solution, since P removal in

sediments has been shown to be lower relative to Al (see section 4.3.4). This was also demonstrated by the achievable reduction of internal P loadings after treatment, in that Al could reduce them by more than a 90% (section 4.3.2, Reitzel et al. (2005)) whereas Phoslock could only reduce them by a maximum of 36% (section 4.3.4).

In terms of pH, Phoslock is effective at a greater pH range than Al, but both include the typical pH levels of lake water, thus, such difference does not become relevant in most lake restoration projects. However, water alkalinity is indeed affected by Al addition but not by Phoslock, consequently widening the latter's application potential (Haghseresht et al. (2009), Afsar and Groves (2009), Gibbs et al. (2011)).

It has been reported that Al increases its P binding uptake after resuspension of sediments (Egemose et al., 2009), but Al can also be released after resuspension (Reitzel et al. (2013), Huser (2012)). By comparison, Phoslock is not capable of fixing P immediately after resuspension (Egemose et al., 2009), but is able to improve sediment consolidation and compaction, allowing rooted vegetation to establish more easily and further decreasing future resuspension events because of increased sediment stabilization (Egemose et al., 2009). Related to that, Al is more effective in removing colour and improving light conditions (Egemose et al., 2009). Therefore, even if Al is more effective in some aspects, Phoslock provides very good sediment properties that can improve the whole sediment-water interface/system and allow the recovery of aquatic vegetation.

In terms of potential toxicity, Phoslock has not been tested enough to say that its ecological risks are controlled (Gibbs et al., 2011), but normal lake water conditions seem to provide a safe environment for Phoslock (and La) (Lürling and Tolman, 2010). As for Al, even with its potential shift to Al^{+3} and $Al(OH)_2^{+}$ at pH 5.5, lake water conditions also provide a safe environment for its application (Afsar and Groves, 2009).

Potential interferences with other compounds, as well as other factors reducing P removal efficiency, seem also greater in Al treatments (see section 2.3.1) than in Phoslock treatments (see section 2.3.2), although this may be due to a better knowledge of Al geochemistry. As mentioned several times, Phoslock is a new product that has not been studied enough, so the absence of interferences could be just due to ignorance. Nevertheless, it would be reasonable to think that interfering factors are shared (i.e., organic matter) since both products bind anionic compounds like phosphate, therefore, this aspect would not be relevant when choosing treatments. The same happens with treatment longevity, which is more or less established for Al only (Welch and Cooke (1999), Huser et al. (2015)).

Regarding recommended or required doses to achieve a determined mobile P reduction, Al doses (see section 2.3.1) are lower than Phoslock's (see section 2.3.2), consequently requiring less mass of the product. In those terms, then, it would be reasonable to assume that Phoslock treatments can become, initially, more expensive. Moreover, comparison of treatment costs also showed that, at equivalent P removal (in %), Phoslock treatments are more expensive than Al treatments (see section 5).

7. Conclusions

Contrary to the hypothesis, there were clear differences in the amount of mobile P reduced in lake sediment via application of Phoslock and Alum. In terms of treatment suitability, Al seems to provide a better treatment for P inactivation from lake sediments than Phoslock due to the more efficient binding under neutral pH conditions. It is reasonable to say that P adsorption with Al should be the preferred treatment for lake and reservoir restoration as long as frequencies of sediment resuspension are low and pH is not above 9, but it might not be as appropriate in shallow lakes. On the other hand, and considering results presented in this thesis, Phoslock treatment could be recommended in wind exposed lakes or lakes with high resuspension frequencies, where water P removal, consolidation of sediments and the reestablishment of lake vegetation would be the most important factors in the recovery process. Moreover, in Phoslock treatments, P already bound to sediments should not present elevated risks for eutrophication and high pH caused by algal blooms due to the low adsorption efficiency of the product. In any case, the product to apply would depend on lake and treatment characteristics, considering all factors involved.

Table 15, Comparison of strengths and weaknesses of treatments for lake restoration.

	Al	Phoslock
P removal effectiveness	✓	×
pH and alkalinity	✓	✓
Sediment properties	×	✓
Toxicity	×	×
Interferences	×	×
Required doses	✓	×
Costs	✓	×

An alternative that could be considered for lake restoration purposes would be to apply a mixture of both Al and Phoslock products, to combine their properties already mentioned in this thesis and achieve maximum P inactivation but also an improvement of lake sediments providing for the recovery of bottom lake vegetation. Another treatment alternative, concerning these two products, could appear from the modification and improvement of the Phoslock compound, maybe with an increased Al content in the clay matrix, allowing a combination of both Al and Phoslock properties in the same product.

In any case, more work needs to be done with Phoslock to fully understand its performance in terms of P adsorption kinetics in water and sediments, adsorption efficiency, treatment longevity and suitability, toxicity and other potential non-target side effects.

Acknowledgements

The author of this thesis is grateful to Brian Huser for the opportunity of learning about this topic, for his role as a supervisor and for his priceless help during the whole project.

The author also thanks Karsten-Raulund Rasmussen (University of Copenhagen) for his role as a co-supervisor of the thesis and his helpful comments.

Thanks also to Grant Douglas (CSIRO) and Nigel Traill (Phoslock Water Solutions Ltd,) for their comments and help regarding Phoslock application and treatment.

Thanks to Jens Fölster and William McNeice for being my examiner and opponent, respectively, and for their helpful comments and review of the thesis.

Thanks to all EnvEuro and other friends I have met during these two years that have helped and motivated me to keep going and be an optimist.

Thanks to my partner, sister and family for all the love you have sent me.

And, finally, thanks to my parents and the EnvEuro program for the opportunity of my life.

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Appendix

A) Water content of sediments

The water content of the sediments for the first and second run was estimated and are displayed in the tables below.

Table 16, Water content of each sediment layer. Sample name indicating sampling point and sample ID, sediment layer, and water content (%wc) are presented. First run.

Name	ID	Layer	%wc
V1_1	1	0-2	94.6
V1_2	2	2-4	92.2
V1_3	3	4-6	92.3
V1_4	4	6-8	91.4
V1_5	5	8-10	90.9
V1_6	6	10-12	90.9
V1_7	7	12-14	90.6
V1_8	8	14-16	90.4
V1_9	9	16-18	89.9
V1_10	10	18-20	90.0
V1_11	11	20-22	89.0
V1_12	12	22-24	89.7
V1_13	13	24-26	89.0
V1_14	14	26-28	88.9
V1_15	15	28-30	88.6
V1_16	16	30-32	87.6
V1_17	17	32-34	87.1
V1_18	18	34-36	86.3

Name	ID	Layer	%wc
V2_1	19	0-2	91.8
V2_2	20	2-4	91.4
V2_3	21	4-6	90.8
V2_4	22	6-8	89.9
V2_5	23	8-10	89.7
V2_6	24	10-12	88.6
V2_7	25	12-14	88.2
V2_8	26	14-16	87.4
V2_9	27	16-18	87.3
V2_10	28	18-20	86.9
V2_11	29	20-22	87.3
V2_12	30	22-24	85.6
V2_13	31	24-26	84.5
V2_14	32	26-28	85.4
V2_15	33	28-30	85.1
V2_16	34	30-32	85.1
V2_17	35	32-34	84.9
V2_18	36	34-36	83.5

Table 17, Water content of each sediment layer. Sample name indicating sampling point and sample ID, sediment layer, and water content (%wc) are presented. Second run.

Name	ID	Layer	wc(%)
V1_1	1	0-2	95.4
V1_2	2	2-4	92.9
V1_3	3	4-6	92.4
V1_4	4	6-8	92.4
V1_5	5	8-10	92.3
V1_6	6	10-12	91.7
V1_7	7	12-14	91.1
V1_8	8	14-16	90.7
V1_9	9	16-18	90.5
V1_10	10	18-20	90.0
V1_11	11	20-22	89.4
V1_12	12	22-24	89.4
V1_13	13	24-26	89.1
V1_14	14	26-28	88.8
V1_15	15	28-30	88.3
V1_16	16	30-32	87.7
V1_17	17	32-34	86.8
V1_18	18	34-36	86.4

Name	ID	Layer	wc(%)
V2_1	19	0-2	92.8
V2_2	20	2-4	91.9
V2_3	21	4-6	91.5
V2_4	22	6-8	90.7
V2_5	23	8-10	89.9
V2_6	24	10-12	89.7
V2_7	25	12-14	89.1
V2_8	26	14-16	92.6
V2_9	27	16-18	88.5
V2_10	28	18-20	88.6
V2_11	29	20-22	87.5
V2_12	30	22-24	86.2
V2_13	31	24-26	85.1
V2_14	32	26-28	85.4
V2_15	33	28-30	85.2
V2_16	34	30-32	85.7
V2_17	35	32-34	84.7
V2_18	36	34-36	83.4

B) Density of sediments

The water content of the sediments for the first and second run was estimated and are displayed in the tables below.

Table 18, Density for each sediment layer. Sample name indicating sampling point and sample ID, sediment layer, and density (g/cm^3) are displayed. Mathematical estimation.

Name	ID	Layer	Density	Name	ID	Layer	Density
V1_1	1	0-2	1.010	V2_1	19	0-2	1.027
V1_2	2	2-4	1.026	V2_2	20	2-4	1.032
V1_3	3	4-6	1.029	V2_3	21	4-6	1.035
V1_4	4	6-8	1.029	V2_4	22	6-8	1.040
V1_5	5	8-10	1.030	V2_5	23	8-10	1.046
V1_6	6	10-12	1.034	V2_6	24	10-12	1.047
V1_7	7	12-14	1.038	V2_7	25	12-14	1.051
V1_8	8	14-16	1.040	V2_8	26	14-16	1.028
V1_9	9	16-18	1.041	V2_9	27	16-18	1.054
V1_10	10	18-20	1.044	V2_10	28	18-20	1.054
V1_11	11	20-22	1.049	V2_11	29	20-22	1.062
V1_12	12	22-24	1.048	V2_12	30	22-24	1.070
V1_13	13	24-26	1.051	V2_13	31	24-26	1.078
V1_14	14	26-28	1.052	V2_14	32	26-28	1.076
V1_15	15	28-30	1.056	V2_15	33	28-30	1.077
V1_16	16	30-32	1.060	V2_16	34	30-32	1.074
V1_17	17	32-34	1.066	V2_17	35	32-34	1.080
V1_18	18	34-36	1.069	V2_18	36	34-36	1.090

Table 19, Density (LOI) for each sediment layer. Sample name indicating sampling point and sample ID, sediment layer, and density (g/cm^3) are displayed.

Name	ID	Layer	Density	Name	ID	Layer	Density
V1_1	1	0-2	1.020	V2_1	19	0-2	1.033
V1_2	2	2-4	1.031	V2_2	20	2-4	1.038
V1_3	3	4-6	1.034	V2_3	21	4-6	1.040
V1_4	4	6-8	1.034	V2_4	22	6-8	1.044
V1_5	5	8-10	1.034	V2_5	23	8-10	1.048
V1_6	6	10-12	1.037	V2_6	24	10-12	1.050
V1_7	7	12-14	1.040	V2_7	25	12-14	1.052
V1_8	8	14-16	1.042	V2_8	26	14-16	1.035
V1_9	9	16-18	1.043	V2_9	27	16-18	1.055
V1_10	10	18-20	1.045	V2_10	28	18-20	1.055
V1_11	11	20-22	1.048	V2_11	29	20-22	1.062
V1_12	12	22-24	1.047	V2_12	30	22-24	1.070
V1_13	13	24-26	1.049	V2_13	31	24-26	1.076
V1_14	14	26-28	1.050	V2_14	32	26-28	1.074
V1_15	15	28-30	1.053	V2_15	33	28-30	1.075
V1_16	16	30-32	1.056	V2_16	34	30-32	1.072
V1_17	17	32-34	1.062	V2_17	35	32-34	1.077
V1_18	18	34-36	1.064	V2_18	36	34-36	1.085

C) P content of fresh sediments

i. Fractionation (first run)

Absorbance (*Table 20*), P content (mg/g, *Table 21*) and mobile P (*Table 22*) are presented.

Table 20, Absorbance values for all P fractions for all 36 analysed samples according to sediment layer and core.

	V1 core					V2 core				
Layer	***Labile***	***Fe-P***	***Al-P***	***Tot-P***	***Ca-P***	***Labile***	***Fe-P***	***Al-P***	***Tot-P***	***Ca-P***
0-2	0.015	0.212	0.074	0.171	0.055	0.010	0.138	0.096	0.204	0.085
2-4	0.017	0.222	0.079	0.178	0.058	0.009	0.163	0.091	0.179	0.076
4-6	0.011	0.266	0.100	0.207	0.074	0.005	0.078	0.062	0.124	0.063
6-8	0.010	0.231	0.076	0.165	0.053	0.005	0.101	0.082	0.151	0.085
8-10	0.009	0.203	0.078	0.173	0.057	0.006	0.127	0.082	0.184	0.080
10-12	0.010	0.395	0.117	0.201	0.058	0.007	0.112	0.078	0.154	0.070
12-14	0.009	0.208	0.069	0.138	0.048	0.008	0.169	0.096	0.172	0.075
14-16	0.012	0.257	0.075	0.146	0.045	0.005	0.063	0.062	0.115	0.062
16-18	0.007	0.142	0.073	0.154	0.053	0.011	0.045	0.066	0.127	0.063
18-20	0.018	0.427	0.136	0.225	0.068	0.050	0.056	0.068	0.120	0.056
20-22	0.006	0.137	0.091	0.189	0.068	0.007	0.092	0.072	0.131	0.060
22-24	0.009	0.169	0.100	0.203	0.068	0.005	0.050	0.059	0.129	0.075
24-26	0.012	0.164	0.078	0.149	0.048	0.006	0.068	0.060	0.130	0.090
26-28	0.006	0.034	0.076	0.141	0.050	0.005	0.026	0.031	0.069	0.063
28-30	0.008	0.054	0.069	0.135	0.044	0.007	0.038	0.047	0.097	0.080
30-32	0.008	0.043	0.074	0.151	0.056	0.005	0.044	0.032	0.068	0.067
32-34	0.007	0.033	0.066	0.122	0.055	0.005	0.048	0.065	0.124	0.072
34-36	0.007	0.036	0.060	0.124	0.054	0.005	0.016	0.021	0.047	0.053

Table 21, P content (mg/g) values for all P fractions for all 36 analysed samples according to sediment layer and core.

	V1 core					V2 core				
Layer	***Labile***	***Fe-P***	***Al-P***	***Tot-P***	***Ca-P***	***Labile***	***Fe-P***	***Al-P***	***Tot-P***	***Ca-P***
0-2	0.039	1.800	0.611	1.459	0.447	0.014	0.741	0.513	1.116	0.452
2-4	0.031	1.268	0.440	1.021	0.318	0.012	0.893	0.493	0.993	0.408
4-6	0.020	1.793	0.662	1.401	0.484	0.003	0.454	0.360	0.747	0.366
6-8	0.020	1.777	0.569	1.273	0.389	0.003	0.483	0.392	0.741	0.407
8-10	0.016	1.487	0.558	1.275	0.401	0.005	0.653	0.418	0.965	0.407
10-12	0.015	2.318	0.674	1.178	0.324	0.008	0.643	0.444	0.903	0.397
12-14	0.015	1.453	0.468	0.966	0.318	0.010	0.924	0.519	0.950	0.401
14-16	0.027	2.018	0.572	1.146	0.332	0.004	0.414	0.412	0.791	0.412
16-18	0.008	0.827	0.418	0.909	0.298	0.019	0.266	0.407	0.810	0.388
18-20	0.037	2.736	0.858	1.440	0.418	0.122	0.343	0.427	0.777	0.348
20-22	0.005	0.689	0.454	0.967	0.335	0.009	0.569	0.444	0.833	0.367
22-24	0.012	0.924	0.541	1.124	0.362	0.002	0.199	0.241	0.548	0.310
24-26	0.026	1.233	0.575	1.130	0.344	0.004	0.263	0.233	0.524	0.356
26-28	0.006	0.195	0.472	0.900	0.303	0.003	0.123	0.155	0.373	0.334
28-30	0.012	0.340	0.446	0.901	0.276	0.006	0.156	0.201	0.435	0.352
30-32	0.009	0.206	0.373	0.785	0.278	0.002	0.181	0.130	0.298	0.288
32-34	0.008	0.174	0.375	0.716	0.309	0.003	0.219	0.307	0.605	0.342
34-36	0.007	0.178	0.315	0.676	0.282	0.003	0.074	0.109	0.275	0.305

Table 22, Mobile P (mg/g) of fresh sediments for all 36 analysed samples according to sediment layer and core. Mobile P includes both labile and Fe-P fractions.

Layer	***Name***	***Mobile P***	***Name***	***Mobile P***
0-2	V1_1	1.839	V2_1	0.755
2-4	V1_2	1.299	V2_2	0.905
4-6	V1_3	1.813	V2_3	0.457
6-8	V1_4	1.797	V2_4	0.486
8-10	V1_5	1.502	V2_5	0.658
10-12	V1_6	2.333	V2_6	0.651
12-14	V1_7	1.468	V2_7	0.933
14-16	V1_8	2.045	V2_8	0.418
16-18	V1_9	0.835	V2_9	0.286
18-20	V1_10	2.774	V2_10	0.466
20-22	V1_11	0.694	V2_11	0.578
22-24	V1_12	0.936	V2_12	0.201
24-26	V1_13	1.259	V2_13	0.267
26-28	V1_14	0.201	V2_14	0.126
28-30	V1_15	0.352	V2_15	0.162
30-32	V1_16	0.215	V2_16	0.184
32-34	V1_17	0.182	V2_17	0.221
34-36	V1_18	0.185	V2_18	0.077

ii. Fractionation (second run)

Fractionation results for the *second run*. Absorbance values and P content (mg/g) are both presented in *Table 23*. Mobile P (sum of labile and Fe-P fractions) is displayed in *Table 24*.

Table 23, Absorbance and P content (mg/g) values for all P fractions for all 24 analysed samples according to sediment layer and core.

		Absorbance		***P content (mg/g)***			***Absorbance***		***P content (mg/g)***	
Layer	***Name***	***Labile***	***Fe-P***	***Labile***	***Fe-P***	***Name***	***Labile***	***Fe-P***	***Labile***	***Fe-P***
0-2	V1_1	NA	NA	NA	NA	V2_1	NA	NA	NA	NA
2-4	V1_2	0.021	0.205	0.047	1.349	V2_2	NA	NA	NA	NA
4-6	V1_3	0.014	0.248	0.025	1.498	V2_3	NA	NA	NA	NA
6-8	V1_4	0.011	0.245	0.018	1.452	V2_4	0.007	0.124	0.007	0.629
8-10	V1_5	0.009	0.279	0.015	1.940	V2_5	0.008	0.155	0.009	0.813
10-12	V1_6	0.011	0.259	0.019	1.675	V2_6	NA	NA	NA	NA
12-14	V1_7	NA	NA	NA	NA	V2_7	0.008	0.135	0.009	0.676
14-16	V1_8	0.014	0.320	0.028	2.113	V2_8	0.007	0.117	0.009	0.758
16-18	V1_9	0.012	0.253	0.022	1.616	V2_9	NA	NA	NA	NA
18-20	V1_10	0.011	0.285	0.016	1.518	V2_10	0.006	0.088	0.004	0.357
20-22	V1_11	0.010	0.302	0.012	1.451	V2_11	0.007	0.092	0.005	0.354
22-24	V1_12	NA	NA	NA	NA	V2_12	NA	NA	NA	NA
24-26	V1_13	NA	NA	NA	NA	V2_13	0.005	0.037	0.002	0.150
26-28	V1_14	0.010	0.091	0.014	0.490	V2_14	0.005	0.043	0.002	0.159
28-30	V1_15	0.012	0.077	0.015	0.330	V2_15	0.006	0.040	0.003	0.127
30-32	V1_16	0.011	0.068	0.012	0.254	V2_16	0.005	0.031	0.002	0.087
32-34	V1_17	NA	NA	NA	NA	V2_17	0.006	0.043	0.004	0.150
34-36	V1_18	NA	NA	NA	NA	V2_18	0.005	0.033	0.003	0.137

** NA ~ Not applicable because not analysed.*

Table 24, Mobile P (mg/g) of fresh sediments for all 24 analysed samples according to sediment layer and core. Mobile P includes both labile and Fe-P fractions. Underlined values correspond to samples that were treated with Al and Phoslock.

Layer	***Name***	***Mobile P***	***Name***	***Mobile P***
0-2	V1_1	NA	V2_1	NA
2-4	V1_2	1.40	V2_2	NA
4-6	V1_3	1.52	V2_3	NA
6-8	V1_4	1.47	V2_4	0.64
8-10	V1_5	1.96	V2_5	0.82
10-12	V1_6	1.69	V2_6	NA
12-14	V1_7	NA	V2_7	0.69
14-16	V1_8	2.14	V2_8	0.77
16-18	V1_9	1.64	V2_9	NA
18-20	V1_10	1.53	V2_10	0.36
20-22	V1_11	1.46	V2_11	0.36
22-24	V1_12	NA	V2_12	NA
24-26	V1_13	NA	V2_13	0.15
26-28	V1_14	0.50	V2_14	0.16
28-30	V1_15	0.35	V2_15	0.13
30-32	V1_16	0.27	V2_16	0.09
32-34	V1_17	NA	V2_17	0.15
34-36	V1_18	NA	V2_18	0.14

** NA ~ Not applicable because not analysed.*

D) P content of sediments after treatment

i. Aluminium

Doses applied to selected sediment samples are displayed in *Table 25*.

Table 25, Al doses (mg/g) applied to sediment samples with different mobile P content (mg/g). Used Al:Mobile P ratios are also indicated (0, 5, 10, 25, 50, 75, 100, 150).

Sample	***Layer***	***Mobile P***	***Ratio / Al (mg/g)***							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	2.14	0.00	10.71	21.41	53.53	107.05	160.58	214.10	321.16
2	20-22	1.46	0.00	7.32	14.63	36.58	73.16	109.74	146.32	219.48
3	8-10	1.96	0.00	9.78	19.55	48.88	97.76	146.63	195.51	293.27
4	4-6	1.52	0.00	7.62	15.23	38.08	76.17	114.25	152.33	228.50
5	2-4	1.40	0.00	6.98	13.95	34.88	69.76	104.64	139.52	209.28
6	12-14	0.69	0.00	3.43	6.85	17.13	34.26	51.40	68.53	102.79
7	18-20	0.36	0.00	1.81	3.61	9.03	18.06	27.09	36.12	54.18
8	28-30	0.13	0.00	0.65	1.31	3.27	6.53	9.80	13.06	19.60

Detailed results for sediment fractionation after Al treatment are presented below. Mobile P including labile and Fe-P fractions, Al-P, Org-P, and Ca-P fractions are determined per each of the *Al:Mobile P* ratios.

Table 26, Remaining Mobile P (mg/g) (including labile and Fe-P) content after Al treatment in sediment samples with different initial mobile P (mg/g). Used Al:Mobile P ratios are also indicated (0, 5, 10, 25, 50, 75, 100, 150).

Sample	***Layer***	***Mobile P***	***Ratios / Mobile P (mg/g)***							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	2.14	1.55	0.87	0.69	0.41	0.25	0.16	0.12	0.08
2	20-22	1.46	1.20	0.68	0.57	0.33	0.21	0.14	0.11	0.07
3	8-10	1.96	1.66	0.80	0.80	0.46	0.30	0.20	0.14	0.08
4	4-6	1.52	1.59	0.81	0.71	0.54	0.27	0.20	0.19	0.11
5	2-4	1.40	1.53	0.99	0.85	0.60	0.41	0.27	0.21	0.16
6	12-14	0.69	0.64	0.40	0.34	0.24	0.18	0.13	0.10	0.09
7	18-20	0.36	0.33	0.22	0.19	0.12	0.10	0.08	0.07	0.05
8	28-30	0.13	0.12	0.12	0.09	0.06	0.05	0.04	0.03	0.02

Table 27, Al-P (mg/g) content after Al treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Al:Mobile P ratios are also indicated (0, 5, 10, 25, 50, 75, 100, 150). NaOH extracts were not centrifuged before analysing, giving higher Al-P values due to organic matter presence.

Sample	***Layer***	***Mobile P***	***Ratios / Al–P (mg/g)***							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	2.14	0.49	1.21	1.43	1.55	1.78	1.84	1.86	1.94
2	20-22	1.46	0.59	1.16	1.26	1.37	1.50	1.58	1.46	1.50
3	8-10	1.96	0.56	1.21	1.52	1.54	1.87	1.82	1.83	1.81
4	4-6	1.52	0.51	1.08	1.24	1.49	1.39	1.56	1.90	1.72
5	2-4	1.40	0.50	1.05	1.23	1.47	1.57	1.64	1.85	1.84
6	12-14	0.69	0.42	0.62	0.71	0.79	0.83	0.87	0.90	0.94
7	18-20	0.36	0.40	0.49	0.54	0.59	0.62	0.64	0.63	0.65
8	28-30	0.13	0.13	0.15	0.17	0.20	0.23	0.23	0.22	0.21

Table 28, Al-P (mg/g) content after Al treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Al:Mobile P ratios are also indicated (0, 5, 10, 25, 50, 75, 100, 150). NaOH extracts were centrifuged before analysing, giving lower Al-P values due to organic matter absence.

Sample	***Layer***	***Mobile P***	***Ratios / Al–P (mg/g)***							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	2.14	0.23	0.89	1.05	1.22	1.40	1.47	1.52	1.66
2	20-22	1.46	0.30	0.82	0.89	1.02	1.12	1.21	1.16	1.29
3	8-10	1.96	0.24	0.81	1.09	1.13	1.42	1.41	1.49	1.51
4	4-6	1.52	0.21	0.69	0.85	1.10	1.01	1.22	1.52	1.44
5	2-4	1.40	0.23	0.73	0.89	1.13	1.24	1.34	1.52	1.54
6	12-14	0.69	0.21	0.39	0.48	0.54	0.59	0.62	0.65	0.70
7	18-20	0.36	0.22	0.30	0.36	0.40	0.42	0.45	0.45	0.48
8	28-30	0.13	0.03	0.04	0.06	0.07	0.08	0.09	0.09	0.09

Table 29, Org-P (mg/g) content after Al treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Al:Mobile P ratios are also indicated (0, 5, 10, 25, 50, 75, 100, 150). NaOH extracts were not centrifuged before analysing, giving lower Org-P values.

Sample	**Layer**	**Mobile P**	**Ratios / Org–P (mg/g)**							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	2.14	0.62	0.51	0.41	0.27	0.25	0.51	0.45	0.27
2	20-22	1.46	0.58	0.39	0.45	0.37	0.42	0.25	0.25	0.58
3	8-10	1.96	0.77	0.71	0.63	0.55	0.28	0.40	0.22	0.24
4	4-6	1.52	0.72	0.69	0.65	0.65	0.47	0.36	0.54	0.44
5	2-4	1.40	0.72	0.91	0.85	0.76	0.74	0.59	0.54	0.60
6	12-14	0.69	0.46	0.59	0.53	0.55	0.52	0.45	0.46	0.48
7	18-20	0.36	0.43	0.45	0.56	0.42	0.45	0.45	0.46	0.44
8	28-30	0.13	0.24	0.27	0.28	0.29	0.28	0.26	0.30	0.32

Table 30, Org-P (mg/g) content after Al treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Al:Mobile P ratios are also indicated (0, 5, 10, 25, 50, 75, 100, 150). NaOH extracts were centrifuged before analysing, giving higher Org-P values.

Sample	**Layer**	**Mobile P**	**Ratios / Org–P (mg/g)**							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	2.14	0.87	0.82	0.79	0.59	0.62	0.88	0.80	0.56
2	20-22	1.46	0.86	0.73	0.82	0.71	0.80	0.62	0.55	0.79
3	8-10	1.96	1.09	1.11	1.06	0.95	0.73	0.81	0.56	0.55
4	4-6	1.52	1.02	1.08	1.04	1.04	0.86	0.70	0.92	0.71
5	2-4	1.40	0.98	1.22	1.19	1.09	1.06	0.88	0.87	0.90
6	12-14	0.69	0.68	0.82	0.76	0.79	0.77	0.70	0.71	0.73
7	18-20	0.36	0.62	0.64	0.75	0.60	0.65	0.64	0.64	0.61
8	28-30	0.13	0.34	0.38	0.39	0.42	0.43	0.39	0.43	0.44

Table 31, Ca-P (mg/g) content after Al treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Al:Mobile P ratios are also indicated (0, 5, 10, 25, 50, 75, 100, 150).

Sample	**Layer**	**Mobile P**	**Ratios / Ca–P (mg/g)**							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	2.14	0.32	0.34	0.34	0.33	0.34	0.36	0.35	0.35
2	20-22	1.46	0.31	0.36	0.35	0.33	0.34	0.32	0.32	0.34
3	8-10	1.96	0.33	0.35	0.33	0.32	0.36	0.34	0.36	0.36
4	4-6	1.52	0.31	0.32	0.34	0.37	0.31	0.32	0.36	0.34
5	2-4	1.40	0.34	0.32	0.33	0.38	0.38	0.34	0.35	0.35
6	12-14	0.69	0.38	0.37	0.39	0.40	0.41	0.39	0.40	0.41
7	18-20	0.36	0.35	0.37	0.37	0.37	0.38	0.37	0.40	0.39
8	28-30	0.13	0.34	0.33	0.34	0.36	0.32	0.31	0.40	0.36

Detailed results of binding efficiency ratios (Al:Al-P) and ratios between Al added and mobile P lost (Al:Mobile P_L) are presented in the tables below.

Table 32, Binding efficiency ratios (Al:Al-P) (by weight) after Al treatment in sediment samples with different initial mobile P (mg/g) concentrations (mobile P values measured at dose ratio 0). Used Al:Mobile P ratios are also indicated (0, 5, 10, 25, 50, 75, 100, 150).

Sample	***Layer***	***Mobile P***	***Ratios Al:Mobile P / Ratios Al:Al-P*** *(by weight)*							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	1.55	0	12	20	44	76	110	141	193
2	20-22	1.20	0	9	16	36	66	91	126	170
3	8-10	1.66	0	12	18	43	69	104	131	194
4	4-6	1.59	0	11	18	35	75	94	100	159
5	2-4	1.53	0	10	16	31	56	78	92	136
6	12-14	0.64	0	9	14	32	58	82	106	148
7	18-20	0.33	0	6	10	23	43	60	81	113
8	28-30	0.12	0	15	24	48	79	106	143	212

Table 33, Al added to mobile P lost ratios (Al:Mobile P_L) (by weight) after Al treatment in sediment samples with different initial mobile P (mg/g) concentrations (mobile P values measured at dose ratio 0). Used Al:Mobile P ratios are also indicated (0, 5, 10, 25, 50, 75, 100, 150).

Sample	***Layer***	***Mobile P***	***Ratios Al:Mobile P / Ratios Al:Mobile P_L*** *(by weight)*							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	1.55	0	16	25	47	82	116	150	219
2	20-22	1.20	0	14	23	42	74	103	133	193
3	8-10	1.66	0	11	23	40	72	100	128	186
4	4-6	1.59	0	10	17	36	58	83	109	155
5	2-4	1.53	0	13	21	38	62	83	106	153
6	12-14	0.64	0	14	23	43	73	100	126	185
7	18-20	0.33	0	16	25	43	78	107	138	192
8	28-30	0.12	0	-263	50	55	95	122	158	213

Detailed results for theoretical and actual binding ratios (in mg P/g Al) are presented in the tables below.

Table 34, Theoretical binding ratios (mg P/g Al) for the 8 treated samples at the Al:Mobile P ratios of 0, 5, 10, 25, 50, 75, 100, and 150. Mobile P values used to calculate the theoretical ratio correspond to measured mobile P at Al dose ratio of 0.

Sample	***Layer***	***Mobile P***	***Ratios Al:Mobile P / Theoretical binding ratios (mg P/g Al)***							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	1.55	0	144.34	72.17	28.87	14.43	9.62	7.22	4.81
2	20-22	1.20	0	164.49	82.25	32.90	16.45	10.97	8.22	5.48
3	8-10	1.66	0	170.24	85.12	34.05	17.02	11.35	8.51	5.67
4	4-6	1.59	0	208.22	104.11	41.64	20.82	13.88	10.41	6.94
5	2-4	1.53	0	218.65	109.33	43.73	21.87	14.58	10.93	7.29
6	12-14	0.64	0	187.84	93.92	37.57	18.78	12.52	9.39	6.26
7	18-20	0.33	0	184.80	92.40	36.96	18.48	12.32	9.24	6.16
8	28-30	0.12	0	177.44	88.72	35.49	17.74	11.83	8.87	5.91

Table 35, Real binding ratios (mg P/g Al) for the 8 treated samples at the Al:Mobile P ratios of 0, 5, 10, 25, 50, 75, 100, and 150. Binding ratios are calculated with P lost and bound to Al.

Sample	***Layer***	***Mobile P***	***Ratios Al:Mobile P / Real binding ratios (mg P/g Al)***							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	1.55	0	63.16	39.86	21.21	12.14	8.62	6.65	4.56
2	20-22	1.20	0	71.75	43.62	23.85	13.51	9.70	7.51	5.18
3	8-10	1.66	0	88.60	44.22	24.72	13.92	10.00	7.78	5.39
4	4-6	1.59	0	102.35	57.60	27.41	17.30	12.12	9.19	6.46
5	2-4	1.53	0	77.38	48.17	26.57	16.04	12.03	9.41	6.52
6	12-14	0.64	0	70.22	44.20	23.32	13.62	10.01	7.93	5.40
7	18-20	0.33	0	64.33	39.50	23.19	12.83	9.38	7.26	5.22
8	28-30	0.12	0	-3.80	20.10	18.31	10.51	8.22	6.33	4.69

Detailed results for achievable internal P loads after treatment are presented in the tables below.

Table 36, Mobile P after treatment (in g/m^2/cm) for the 8 treated samples at the Al:Mobile P ratios of 0, 5, 10, 25, 50, 75, 100, and 150. Mobile P values in mg/g correspond to measured mobile P at Al dose ratio of 0.

Sample	***Layer***	***Mobile P***	***Ratios Al:Mobile P / Mobile P (g/m^2/cm)***							
	(cm)	*(mg/g)*	0	5	10	25	50	75	100	150
1	14-16	1.55	1.49	0.84	0.67	0.40	0.24	0.16	0.12	0.08
2	20-22	1.20	1.34	0.75	0.63	0.37	0.24	0.15	0.12	0.08
3	8-10	1.66	1.32	0.63	0.63	0.36	0.24	0.16	0.11	0.07
4	4-6	1.59	1.25	0.63	0.56	0.43	0.21	0.16	0.15	0.09
5	2-4	1.53	1.11	0.72	0.62	0.44	0.30	0.19	0.15	0.12
6	12-14	0.64	0.74	0.46	0.39	0.28	0.20	0.15	0.12	0.10
7	18-20	0.33	0.40	0.26	0.23	0.15	0.12	0.10	0.09	0.06
8	28-30	0.12	0.19	0.19	0.14	0.09	0.08	0.06	0.05	0.04

Table 37, Achievable internal P loads (Li, in mg/m^2/d) after treatment for the 8 treated samples at the Al:Mobile P ratios of 0, 5, 10, 25, 50, 75, 100, and 150. Maximum achievable reduction of Li (in %) is also indicated.

Sample	***Layer***	***Li reduction***	***Ratios Al:Mobile P / Li (mg/m^2/d)***							
	(cm)	*(maximum)*	0	5	10	25	50	75	100	150
1	14-16	98%	21.78	11.94	9.36	5.27	2.88	1.64	1.07	0.47
2	20-22	98%	19.51	10.70	8.79	4.86	2.91	1.64	1.07	0.43
3	8-10	98%	19.25	8.87	8.89	4.77	2.93	1.68	1.01	0.31
4	4-6	97%	18.14	8.88	7.72	5.74	2.49	1.70	1.51	0.61
5	2-4	93%	16.12	10.17	8.71	5.90	3.78	2.23	1.64	1.08
6	12-14	92%	10.44	6.27	5.20	3.52	2.36	1.53	1.04	0.82
7	18-20	96%	5.37	3.25	2.77	1.56	1.16	0.75	0.60	0.23
8	28-30	106%	2.10	2.16	1.47	0.66	0.44	0.15	0.10	-0.12

ii. Phoslock (first addition)

Doses applied to selected sediment samples are displayed in *Table 38*.

Table 38, Phoslock doses (mg/g) applied to sediment samples with different mobile P content (mg/g). Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios / Phoslock (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	0.00	32.12	64.23	160.58	321.16	481.73	642.31	963.47
2	20-22	1.46	0.00	21.95	43.90	109.74	219.48	329.21	438.95	658.43
3	8-10	1.96	0.00	29.33	58.65	146.63	293.27	439.90	586.53	879.80
4	4-6	1.52	0.00	22.85	45.70	114.25	228.50	342.75	456.99	685.49
5	2-4	1.40	0.00	20.93	41.86	104.64	209.28	313.91	418.55	627.83
6	12-14	0.69	0.00	10.28	20.56	51.40	102.79	154.19	205.58	308.38
7	18-20	0.36	0.00	5.42	10.84	27.09	54.18	81.27	108.36	162.54
8	28-30	0.13	0.00	1.96	3.92	9.80	19.60	29.39	39.19	58.79

Detailed results for sediment fractionation after Phoslock treatment are presented below. Mobile P including labile and Fe-P fractions, Al-P, Org-P, and Ca-P fractions are determined per each of the *Phoslock:Mobile P* ratios.

Table 39, Remaining Mobile P (mg/g) (labile and Fe-P) content after Phoslock treatment in sediment samples with different initial mobile P (mg/g). Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios / Mobile P (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	1.63	1.61	1.56	1.40	1.08	1.11	1.05	1.16
2	20-22	1.46	1.16	1.15	1.18	1.07	0.87	0.75	0.72	0.83
3	8-10	1.96	1.56	1.54	1.54	1.44	1.21	1.08	1.17	1.20
4	4-6	1.52	1.59	1.59	1.54	1.44	1.32	1.17	1.15	1.13
5	2-4	1.40	1.56	1.50	1.55	1.37	1.62	0.96	1.03	0.90
6	12-14	0.69	0.61	0.58	0.58	0.67	0.51	0.41	0.41	0.37
7	18-20	0.36	0.43	0.56	0.30	0.28	0.41	0.46	0.26	0.18
8	28-30	0.13	0.09	0.14	0.17	0.11	0.02	0.26	0.23	0.00

Table 40, Al-P (mg/g) content after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios / Al–P (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	0.45	0.44	0.49	0.53	0.53	0.55	0.49	0.43
2	20-22	1.46	0.50	0.51	0.52	0.54	0.54	0.53	0.51	0.45
3	8-10	1.96	0.42	0.43	0.44	0.48	0.52	0.52	0.49	0.44
4	4-6	1.52	0.42	0.45	0.44	0.48	0.53	0.53	0.52	0.45
5	2-4	1.40	0.32	0.23	0.26	0.38	0.53	0.95	0.84	0.47
6	12-14	0.69	0.24	0.34	0.25	0.28	0.53	0.29	0.31	0.35
7	18-20	0.36	0.18	0.27	0.44	0.24	0.28	0.31	0.46	0.24
8	28-30	0.13	0.05	0.13	0.08	0.07	0.06	0.10	0.16	0.09

Table 41, Org-P (mg/g) content after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios / Org–P (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	0.83	0.85	1.00	1.14	1.08	0.98	0.87	0.85
2	20-22	1.46	0.69	1.08	0.74	0.74	0.67	0.61	0.66	0.71
3	8-10	1.96	0.80	0.89	1.06	1.04	1.01	0.91	1.04	0.99
4	4-6	1.52	0.86	0.87	0.89	0.98	0.93	0.86	1.06	1.02
5	2-4	1.40	0.74	1.20	0.93	0.70	0.62	0.56	0.61	0.67
6	12-14	0.69	0.58	0.45	0.52	0.60	0.55	0.53	0.63	0.50
7	18-20	0.36	0.52	0.52	0.38	0.65	0.57	0.74	0.41	0.56
8	28-30	0.13	0.47	0.46	0.22	0.33	0.41	0.26	0.16	0.22

Table 42, Ca-P (mg/g) content after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios / Ca–P (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	0.40	0.44	0.70	0.70	1.29	1.37	1.21	1.29
2	20-22	1.46	0.36	0.36	0.42	0.57	0.79	1.15	1.00	1.24
3	8-10	1.96	0.39	0.44	0.48	0.55	0.82	1.06	1.43	1.19
4	4-6	1.52	0.35	0.42	0.45	0.49	0.75	0.86	1.05	1.41
5	2-4	1.40	0.14	0.09	0.09	0.27	0.59	0.87	0.83	0.78
6	12-14	0.69	0.32	0.24	0.42	0.23	0.36	0.41	0.52	0.64
7	18-20	0.36	0.27	0.22	0.26	0.19	0.49	0.37	0.31	0.48
8	28-30	0.13	0.26	0.25	0.18	0.43	0.24	0.25	0.27	0.25

Detailed results of binding efficiency ratios (Phoslock:Al-P and Phoslock:Ca-P) and ratios between Phoslock added and mobile P lost (Phoslock:Mobile P_L) are presented in the tables below.

Table 43, Binding efficiency ratios (Phoslock:Al-P) (by weight) after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations (mobile P values measured at dose ratio 0). Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios Phoslock:Mobile P / Ratios Phoslock:Al-P*** *(by weight)*							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	1.63	0	73	132	305	604	879	1305	2220
2	20-22	1.16	0	43	85	203	405	623	854	1466
3	8-10	1.56	0	68	132	306	566	843	1191	2022
4	4-6	1.59	0	51	103	239	432	650	887	1516
5	2-4	1.56	0	90	159	277	396	330	498	1330
6	12-14	0.61	0	30	82	186	192	536	656	888
7	18-20	0.43	0	20	25	112	195	258	237	669
8	28-30	0.09	0	15	50	143	319	300	243	674

Table 44, Binding efficiency ratios (Phoslock:Ca-P) (by weight) after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations (mobile P values measured at dose ratio 0). Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios Phoslock:Mobile P / Ratios Phoslock:Ca-P*** *(by weight)*							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	1.63	0	80	146	230	456	374	470	799
2	20-22	1.16	0	61	121	264	384	418	383	656
3	8-10	1.56	0	75	132	307	531	538	552	616
4	4-6	1.59	0	65	110	256	470	459	534	653
5	2-4	1.56	0	155	482	1113	790	533	481	760
6	12-14	0.61	0	32	84	121	446	430	502	592
7	18-20	0.43	0	20	50	105	281	164	291	527
8	28-30	0.09	0	8	15	54	46	121	159	218

Table 45, Phoslock added to mobile P lost ratios (Phoslock:Mobile P_L) (by weight) after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations (mobile P values measured at dose ratio 0). Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios Phoslock:Mobile P / Phoslock:Mobile P_L*** *(by weight)*							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	1.63	0	1210	886	683	577	918	1103	2037
2	20-22	1.16	0	1417	-3123	1138	754	791	986	1979
3	8-10	1.56	0	1936	2914	1229	851	925	1498	2450
4	4-6	1.59	0	11626	965	771	843	813	1052	1503
5	2-4	1.56	0	331	5040	547	-3549	525	788	944
6	12-14	0.61	0	359	583	-800	1036	761	1052	1272
7	18-20	0.43	0	-41	81	175	3038	-2643	615	647
8	28-30	0.09	0	-45	-54	-880	252	-182	-292	629

Detailed results for theoretical and actual binding ratios (in mg P/g Phoslock) are presented in the tables below.

Table 46, Theoretical binding ratios (mg P/g Phoslock) for the 8 treated samples at the Phoslock:Mobile P ratios of 0, 15, 30, 75, 150, 225, 300, and 450. Mobile P values used to calculate the theoretical ratio correspond to measured mobile P at Phoslock dose ratio of 0.

Sample	***Layer***	***Mobile P***	***Phoslock:Mobile P / Theoretical binding ratio (mg P/g Phoslock)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	1.63	0	50.86	25.43	10.17	5.09	3.39	2.54	1.70
2	20-22	1.16	0	53.00	26.50	10.60	5.30	3.53	2.65	1.77
3	8-10	1.56	0	53.11	26.55	10.62	5.31	3.54	2.66	1.77
4	4-6	1.59	0	69.52	34.76	13.90	6.95	4.63	3.48	2.32
5	2-4	1.56	0	74.68	37.34	14.94	7.47	4.98	3.73	2.49
6	12-14	0.61	0	59.37	29.69	11.87	5.94	3.96	2.97	1.98
7	18-20	0.43	0	79.77	39.89	15.95	7.98	5.32	3.99	2.66
8	28-30	0.09	0	48.34	24.17	9.67	4.83	3.22	2.42	1.61

Table 47, Real binding ratios (mg P/g Phoslock) for the 8 treated samples at the Phoslock:Mobile P ratios of 0, 15, 30, 75, 150, 225, 300, and 450. Binding ratios are calculated with P lost and bound to Phoslock.

Sample	***Layer***	***Mobile P***	***Phoslock:Mobile P / Real binding ratio (mg P/g Phoslock)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	1.63	0	0.83	1.13	1.46	1.73	1.09	0.91	0.49
2	20-22	1.16	0	0.71	-0.32	0.88	1.33	1.26	1.01	0.51
3	8-10	1.56	0	0.52	0.34	0.81	1.18	1.08	0.67	0.41
4	4-6	1.59	0	0.09	1.04	1.30	1.19	1.23	0.95	0.67
5	2-4	1.56	0	3.02	0.20	1.83	-0.28	1.90	1.27	1.06
6	12-14	0.61	0	2.78	1.71	-1.25	0.97	1.31	0.95	0.79
7	18-20	0.43	0	-24.25	12.38	5.70	0.33	-0.38	1.63	1.54
8	28-30	0.09	0	-22.25	-18.37	-1.14	3.96	-5.50	-3.43	1.59

Detailed results for achievable internal P loads after treatment are presented in the tables below.

Table 48, Mobile P after treatment (in g/m^2/cm) for the 8 treated samples at the Phoslock:Mobile P ratios of 0, 15, 30, 75, 150, 225, 300, and 450. Mobile P values in mg/g correspond to measured mobile P at Phoslock dose ratio of 0.

Sample	***Layer***	***Mobile P***	***Ratios Phoslock:Mobile P / Mobile P (g/m^2/cm)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	1.63	1.57	1.55	1.50	1.35	1.04	1.07	1.01	1.12
2	20-22	1.16	1.29	1.28	1.31	1.19	0.97	0.83	0.80	0.92
3	8-10	1.56	1.24	1.22	1.22	1.14	0.96	0.86	0.93	0.95
4	4-6	1.59	1.25	1.25	1.21	1.13	1.04	0.92	0.91	0.89
5	2-4	1.56	1.14	1.10	1.14	1.00	1.18	0.70	0.75	0.66
6	12-14	0.61	0.70	0.67	0.66	0.77	0.59	0.47	0.48	0.42
7	18-20	0.43	0.52	0.68	0.36	0.33	0.50	0.56	0.31	0.22
8	28-30	0.09	0.15	0.22	0.27	0.17	0.03	0.41	0.37	0.00

Table 49, Achievable internal P loads (Li, in mg/m^2/d) after treatment for the 8 treated samples at the Phoslock:Mobile P ratios of 0, 15, 30, 75, 150, 225, 300, and 450. Maximum achievable reduction of Li (in %) is also indicated (with the 225 dose).

Sample	***Layer***	***Li reduction***	***Ratios Phoslock:Mobile P / Li (mg/m^2/d)***							
	(cm)	*(maximum)*	0	15	30	75	150	225	300	450
1	14-16	33%	23.06	22.67	22.00	19.64	14.96	15.43	14.59	16.18
2	20-22	37%	18.84	18.58	19.07	17.22	13.94	11.84	11.36	13.25
3	8-10	32%	17.98	17.79	17.73	16.54	13.84	12.27	13.28	13.67
4	4-6	28%	18.17	18.15	17.61	16.41	14.96	13.16	13.01	12.75
5	2-4	40%	16.54	15.84	16.44	14.43	17.19	9.94	10.68	9.20
6	12-14	36%	9.86	9.37	9.25	10.97	8.14	6.36	6.48	5.67
7	18-20	-8%	7.16	9.54	4.72	4.35	6.83	7.71	3.95	2.59
8	28-30	-246%	1.59	2.64	3.33	1.86	-0.29	5.49	4.83	-0.67

iii. Phoslock (second addition)

Doses applied to selected sediment samples are displayed in *Table 50*.

Table 50, Phoslock doses (mg/g) applied to sediment samples with different mobile P content (mg/g). Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios / Phoslock (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	0.00	32.12	64.23	160.58	321.16	481.73	642.31	963.47
2	20-22	1.46	0.00	21.95	43.90	109.74	219.48	329.21	438.95	658.43
3	8-10	1.96	0.00	29.33	58.65	146.63	293.27	439.90	586.53	879.80
4	4-6	1.52	0.00	22.85	45.70	114.25	228.50	342.75	456.99	685.49

Detailed results for sediment fractionation after Phoslock treatment are presented below. Mobile P including labile and Fe-P fractions, Al-P, Org-P, and Ca-P fractions are determined per each of the Al:Mobile P ratios.

Table 51, Remaining Mobile P (mg/g) (labile and Fe-P) content after Phoslock treatment in sediment samples with different initial mobile P (mg/g). Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios / Mobile P (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	2.07	2.07	2.18	1.70	1.35	1.47	1.52	1.62
2	20-22	1.46	1.51	1.47	1.47	1.33	1.13	1.16	1.21	1.26
3	8-10	1.96	2.05	2.06	2.03	1.97	1.50	1.60	1.64	1.81
4	4-6	1.52	1.94	1.99	1.98	1.94	1.65	1.68	1.47	1.63

Table 52, Al-P (mg/g) content after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450). NaOH extracts were not centrifuged before analysing, giving higher Al-P values due to organic matter presence.

Sample	***Layer***	***Mobile P***	***Ratios / Al–P (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	0.33	0.34	0.40	0.43	0.43	0.43	0.40	0.34
2	20-22	1.46	0.41	0.41	0.42	0.46	0.48	0.45	0.44	0.41
3	8-10	1.96	0.33	0.34	0.33	0.39	0.45	0.44	0.40	0.33
4	4-6	1.52	0.32	0.33	0.34	0.36	0.43	0.38	0.39	0.32

Table 53, Al-P (mg/g) content after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450). NaOH extracts were centrifuged before analysing, giving lower Al-P values due to organic matter absence.

Sample	***Layer***	***Mobile P***	***Ratios / Al–P (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	0.11	0.13	0.18	0.26	0.30	0.31	0.29	0.25
2	20-22	1.46	0.17	0.20	0.20	0.28	0.34	0.33	0.32	0.30
3	8-10	1.96	0.10	0.12	0.12	0.18	0.31	0.31	0.30	0.24
4	4-6	1.52	0.09	0.10	0.11	0.14	0.27	0.25	0.28	0.23

Table 54, Org-P (mg/g) content after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450). NaOH extracts were not centrifuged before analysing, giving lower Org-P values.

Sample	***Layer***	***Mobile P***	***Ratios / Org–P (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	0.57	0.54	0.60	0.55	0.57	0.59	0.56	0.55
2	20-22	1.46	0.53	0.51	0.53	0.54	0.51	0.52	0.52	0.53
3	8-10	1.96	0.65	0.63	0.64	0.65	0.66	0.65	0.62	0.64
4	4-6	1.52	0.59	0.61	0.63	0.62	0.63	0.63	0.66	0.62

Table 55, Org-P (mg/g) content after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450). NaOH extracts were centrifuged before analysing, giving higher Org-P values.

Sample	***Layer***	***Mobile P***	***Ratios / Org–P (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	0.80	0.75	0.83	0.72	0.70	0.71	0.67	0.64
2	20-22	1.46	0.77	0.73	0.75	0.72	0.65	0.64	0.63	0.64
3	8-10	1.96	0.89	0.85	0.86	0.85	0.79	0.78	0.73	0.73
4	4-6	1.52	0.82	0.84	0.85	0.84	0.80	0.76	0.76	0.70

Table 56, Ca-P (mg/g) content after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations. Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios / Ca–P (mg/g)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.14	0.32	0.35	0.38	0.64	0.93	0.94	0.95	0.92
2	20-22	1.46	0.32	0.32	0.34	0.45	0.67	0.66	0.68	0.73
3	8-10	1.96	0.31	0.33	0.34	0.47	0.85	0.81	0.85	0.81
4	4-6	1.52	0.30	0.32	0.35	0.38	0.70	0.71	0.87	0.80

Detailed results of binding efficiency ratios (Phoslock:Al-P and Phoslock:Ca-P) and ratios between Phoslock added and mobile P lost (Phoslock:Mobile P_L) are presented in the tables below.

Table 57, Binding efficiency ratios (Phoslock:Al-P) (by weight) after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations (mobile P values measured at dose ratio 0). Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios Phoslock:Mobile P / Ratios Phoslock:Al-P*** *(by weight)*							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.07	0	246	358	621	1065	1546	2178	3818
2	20-22	1.51	0	112	217	395	641	987	1354	2171
3	8-10	2.05	0	254	491	803	938	1437	1974	3626
4	4-6	1.94	0	220	399	817	861	1371	1615	2931

Table 58, Binding efficiency ratios (Phoslock:Ca-P) (by weight) after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations (mobile P values measured at dose ratio 0). Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios Phoslock:Mobile P / Ratios Phoslock:Ca-P*** *(by weight)*							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.07	0	91	168	252	346	512	675	1051
2	20-22	1.51	0	68	128	242	325	495	649	897
3	8-10	2.05	0	88	171	311	345	544	693	1088
4	4-6	1.94	0	72	130	304	325	483	523	852

Table 59, Phoslock added to mobile P lost ratios (Phoslock:Mobile P_L) (by weight) after Phoslock treatment in sediment samples with different initial mobile P (mg/g) concentrations (mobile P values measured at dose ratio 0). Used Phoslock:Mobile P ratios are also indicated (0, 15, 30, 75, 150, 225, 300, 450).

Sample	***Layer***	***Mobile P***	***Ratios Phoslock:Mobile P / Phoslock:Mobile P_L*** *(by weight)*							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.07	0	22320	-581	438	447	802	1179	2130
2	20-22	1.51	0	636	1287	635	579	943	1475	2659
3	8-10	2.05	0	-2367	3340	2029	540	981	1430	3675
4	4-6	1.94	0	-496	-1214	27308	791	1299	970	2184

Detailed results for theoretical and actual binding ratios (in mg P/g Phoslock) are presented in the tables below.

Table 60, Theoretical binding ratios (mg P/g Phoslock) for the 4 treated samples at the Phoslock:Mobile P ratios of 0, 15, 30, 75, 150, 225, 300, and 450. Mobile P values used to calculate the theoretical ratio correspond to measured mobile P at Phoslock dose ratio of 0.

Sample	***Layer***	***Mobile P***	***Phoslock:Mobile P / Theoretical binding ratio (mg P/g Phoslock)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.07	0	64.40	32.20	12.88	6.44	4.29	3.22	2.15
2	20-22	1.51	0	68.61	34.31	13.72	6.86	4.57	3.43	2.29
3	8-10	2.05	0	69.77	34.88	13.95	6.98	4.65	3.49	2.33
4	4-6	1.94	0	85.04	42.52	17.01	8.50	5.67	4.25	2.83

Table 61, Real binding ratios (mg P/g Phoslock) for the 4 treated samples at the Phoslock:Mobile P ratios of 0, 15, 30, 75, 150, 225, 300, and 450. Binding ratios are calculated with P lost and bound to Phoslock.

Sample	***Layer***	***Mobile P***	***Phoslock:Mobile P / Real binding ratio (mg P/g Phoslock)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.07	0	0.04	-1.72	2.28	2.24	1.25	0.85	0.47
2	20-22	1.51	0	1.57	0.78	1.57	1.73	1.06	0.68	0.38
3	8-10	2.05	0	-0.42	0.30	0.49	1.85	1.02	0.70	0.27
4	4-6	1.94	0	-2.02	-0.82	0.04	1.26	0.77	1.03	0.46

Detailed results for achievable internal P loads after treatment are presented in the tables below.

Table 62, Mobile P after treatment (in g/m^2/cm) for the 4 treated samples at the Phoslock:Mobile P ratios of 0, 15, 30, 75, 150, 225, 300, and 450. Mobile P values in mg/g correspond to measured mobile P at Phoslock dose ratio of 0.

Sample	***Layer***	***Mobile P***	***Ratios Phoslock:Mobile P / Mobile P (g/m^2/cm)***							
	(cm)	*(mg/g)*	0	15	30	75	150	225	300	450
1	14-16	2.07	1.99	1.99	2.10	1.64	1.30	1.41	1.47	1.56
2	20-22	1.51	1.68	1.64	1.64	1.48	1.25	1.29	1.34	1.40
3	8-10	2.05	1.62	1.63	1.61	1.57	1.19	1.27	1.30	1.43
4	4-6	1.94	1.53	1.57	1.56	1.53	1.30	1.32	1.16	1.28

Table 63, Achievable internal P loads (Li, in mg/m^2/d) after treatment for the 4 treated samples at the Phoslock:Mobile P ratios of 0, 15, 30, 75, 150, 225, 300, and 450. Maximum achievable reduction of Li (in %) is also indicated (at the 150 dose).

Sample	***Layer***	***Li reduction***	***Ratios Phoslock:Mobile P / Li (mg/m^2/d)***							
	(cm)	*(maximum)*	0	15	30	75	150	225	300	450
1	14-16	36%	29.38	29.36	30.99	24.05	18.93	20.64	21.46	22.80
2	20-22	26%	24.59	24.01	24.02	21.69	18.23	18.73	19.59	20.43
3	8-10	27%	23.83	23.98	23.62	22.97	17.32	18.46	18.91	20.96
4	4-6	15%	22.39	22.94	22.84	22.34	18.96	19.25	16.79	18.66